

Thermophysical Properties and Phase Behavior of Supercritical Fluids

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Bachelor's thesis

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17.5.2026

Turku

Bachelor's thesis

Subject: Mechanical Engineering

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Title: Thermophysical Properties and Phase Behavior of Supercritical Fluids

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Number of pages: 21 pages

Date: 17.5.2026

Abstract

Supercritical fluids have attracted increasing scientific interest because of their unique combination of liquid-like and gas-like properties depending on temperature and pressure. When a substance exceeds its critical temperature and pressure, it enters to a supercritical state in which the clear interface between liquid and gas phases disappears. This state is classified as a single phase and has complex and highly variable behavior.

This work presents an overview of the phase behavior and thermophysical properties of supercritical fluids. Focus is placed on the behavior near the critical point and pseudo-critical region, where properties such as density and specific heat capacity change strongly and nonlinear. The Widom line and pseudo-boiling are discussed as key tools for describing continuous transitions from liquid-like to gas-like or vice versa in the supercritical region.

Understanding phase behavior and thermophysical properties of supercritical fluids is essential for theoretical analysis and engineering applications. Because of the complexity of the supercritical state, it remains an active area of research in thermodynamics and fluid mechanics.

Keywords: Pseudo-boiling, Supercritical fluid, Widom line

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1 Introduction

Supercritical fluids (SCFs) have attracted increasing scientific interest in recent decades because of their unusual physical properties. A substance becomes supercritical when its temperature and pressure exceed the critical point. Above the critical point normal phase boundary between liquid and gas disappears, this means that the substance can have combined properties of liquid and gas. However supercritical fluid is not separate gas and liquid phases, instead the fluid is single supercritical phase. For example, supercritical fluids can have densities like liquid while it maintains viscosity and diffusivity like gas, this makes supercritical fluids very interesting in thermodynamics and heat-transfer. Supercritical fluid with these properties makes it very useful for energy systems, chemical processing and extraction technologies. [1]

Near and above the critical point, thermophysical properties such as density, heat capacity, and compressibility can change significantly. In classical thermodynamics, it is normally assumed that above the critical point fluid appears as a single phase, where gas-like and liquid-like behavior can't be recognized. However, more recent studies have shown that the supercritical region is not fully uniform, but different regions of behavior can be detected in it. Between these regions there is no true phase transition, but transitions occur as continuous crossovers in thermodynamic properties. [2], [3]

One key concept of describing these phenomena is the Widom line, which is defined by a set of points that are maximum values of thermodynamic properties such as heat capacity and compressibility. This line is used to describe the transition between liquid-like and gas-like behavior in supercritical fluid. Widom line offers a good way to analyze how thermophysical properties are changing in supercritical fluids. [2]

Afterwards have found that supercritical fluids can contain multiple crossover lines which separate liquid-like and gas-like behavior. These different models give more perspective to analyze supercritical state. However, there is not a simple way to find clear interface between liquid-like and gas-like. That is why supercritical state remains an active area of research in physics and thermodynamics.[2], [3]

This work examines the thermodynamic behavior and thermophysical properties of supercritical fluids. Chapter 2 introduces the fundamental concepts of phase behavior, phase diagrams, and critical phenomena. Chapter 3 focuses on the physical interpretation of the supercritical state, including the Widom line and pseudo-boiling. Chapter 4 discusses the most important

thermophysical properties of supercritical fluids, such as density, compressibility, specific heat capacity, and viscosity. Finally, Chapter 5 presents engineering and industrial applications of supercritical fluids, including supercritical power cycles and extraction technologies.

Understanding these phenomena and modelling types are necessary to accurately determine thermophysical properties and phase behavior of supercritical fluids. The objective of this work is to give an easily understandable overview of these phenomena.

Artificial intelligence was used as a tool to assist with translation and proofreading. The AI helped to improve language clarity, grammar, and overall readability; however the scientific content and ideas remain my own.

2 Phase behavior and critical phenomena

2.1 Phases of matter

A substance can appear as different physical states depending on temperature and pressure. The most common states are solid, liquid and gas. Those three states differ in the arrangement and mobility of molecules, as shown in Figure 1. In solid phase molecules are arranged in ordered structure and the movement of molecules is very limited. In liquids molecules are still close together but they can flow past each other. That is why liquids can flow but maintain constant volume. Third well known phase is gas, where distance between molecules is much longer and movement is much faster. This leads to low density and high compressibility. [4]

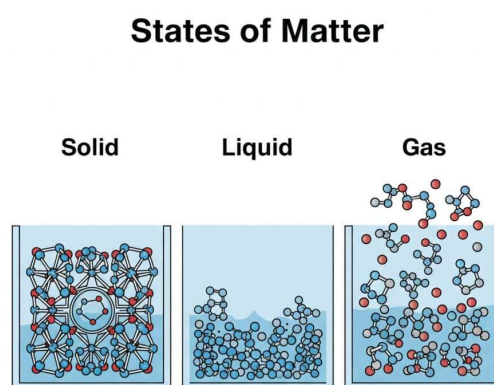


Figure 1: Difference in molecular motion in different states of matter. [5]

The phase of a substance is determined by forces between molecules and thermal energy of molecules. When temperatures are low and pressures are high, forces between molecules dominate and then the substance usually appears as compressed like solid or liquid. When temperature rises the kinetic energy of molecules grows and overcomes the forces between molecules. [4] This leads to phase transitions such as melting or vaporization. Movement between molecules can be seen simplified in Figure 1. These phase transitions are usually described by phase diagrams, which show the stable phase of a substance with a function of temperature and pressure. [6]

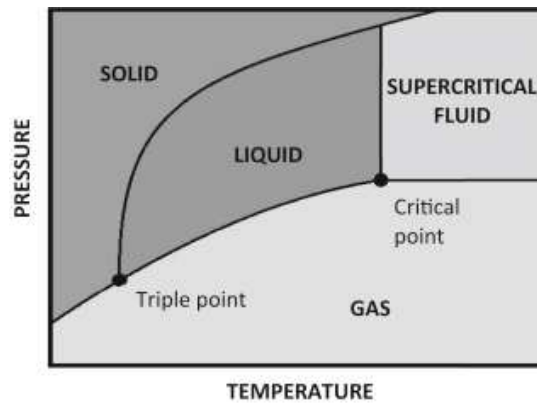


Figure 2: Typical two-dimensional phase diagram including the supercritical region. [1]

In addition to the classical phases, fluids can also exist in conditions where there is no clear difference between liquid and gas. These conditions bring matter into a supercritical state, as shown in Figure 2. Near critical point thermophysical properties can change rapidly even with small changes in temperature or pressure. These changes are caused by large variations in density and energy inside the fluid. [7]

As mentioned earlier, above the critical point the clear interface between liquid and gas disappears and a substance forms a single phase and that is called supercritical fluid. This phase has become important in modern energy and thermal systems. Studies of turbulent and heat-transfer processes under these conditions show that large rises in thermophysical properties can significantly affect flow structures and thermal boundary layers. Understanding the different phases of matter and the phase transitions is essential when studying supercritical fluids. The behavior of fluids near the critical point provides the foundation for analyzing the thermophysical properties and phase behavior. [8]

2.2 Phase diagrams and the critical point

Phase diagrams are essential tools in thermo- and fluid mechanics. Phase diagrams show physical state of a substance and can be described by the function of temperature and pressure. In a typical pressure-temperature phase diagram in Figure 2, different sections correspond to stable phase and boundaries between sections present a state where two phases are in equilibrium. These boundaries define phase transitions like melting or vaporization and provide a framework for understanding how substances change phase under different conditions.

The liquid-vapor coexistence curve is one of the most important things in phase diagrams. Along this curve, the difference between the liquid and gas phases gradually decreases. In the

end of the curve the densities of two phases become as one, this is a combination of temperature and pressure known as critical point. [9]

The concept of the critical point was established in nineteenth century with experiments of carbon dioxide, where it was proven that after certain point gas cannot be condensed with only increasing pressure. These findings led to an idea of continuity of liquid and gas states and later to theoretical descriptions of critical behavior in fluids. In today's thermodynamics the critical point is interpreted as a limit of the liquid-gas equilibrium where thermophysical properties can change rapidly. [10]

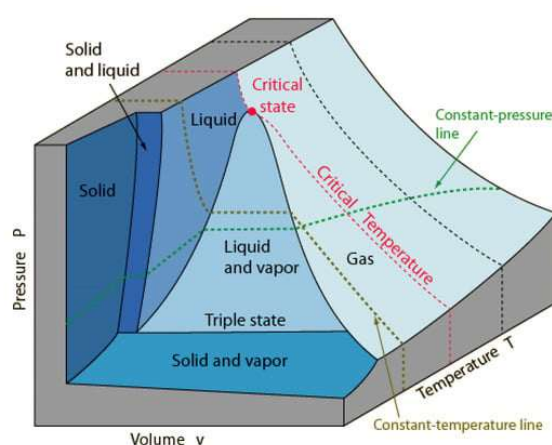


Figure 3: Three-dimensional phase diagram shows more accurately the definition of critical point. [10]

When temperature and pressure go above critical point a substance enters supercritical region seen in Figure 3. In supercritical regions the physical state of matter cannot be described as a function of temperature and pressure anymore. However, recent studies have shown that even in supercritical region different behavior zones can be identified. For that we have these crossover lines mentioned earlier like Widom-, Frenkel, and pseudo-critical line. [10]

2.3 Behavior near the critical point

When a fluid approaches the critical point, its behavior changes even more complex and it cannot be described with simple distinctions between liquid and gas phases. Instead of clear phase transition there are continuous and strongly nonlinear changes in thermodynamical and transport properties. One of the key features of this region is large density fluctuations which are due to the increased length between molecules. These changes affect strongly to macroscopic properties and lead to very different behavior compared to typical fluids. Whenever talking

about supercritical phase, should be very clear that there is no real phase change after reaching the critical point. [10]

Near the critical point many thermodynamical response functions reach their maximum value. For example, isothermal compressibility and isobaric heat capacity can increase quickly because of the fluctuations in energy and density. These peak values are not just indicators of supercritical behavior, but they play a key role in defining crossover lines in the supercritical region. [3]

Also transport properties such as viscosity and thermal conductivity can behave in unusual way near the critical point. These changes are very important in supercritical flows, where strong increases in density and heat capacity can affect turbulence structures and thermal boundary layers. Recent studies have shown that supercritical conditions can have significant impact on flow and heat transfer efficiency. [11]

3 Supercritical fluids

3.1 Physical interpretation of the supercritical state

The supercritical state can be interpreted as a thermodynamic region, where the normal interface between liquid and gas cannot be recognized. The fluid does not become fully homogeneous. Instead, there are continuous variations in density and molecular organization. Studies have shown that above critical point fluids can have heterogeneous structures, where liquid-like molecule clusters can appear in gas-like environment. These structures form and break down dynamically. [10]

Fluids near the pseudo-critical region can have strong density and energy variations. Under subcritical two-phase conditions, liquid and gas phases can coexist and are separated by a clear interface. So, the fluid cannot be described as fully homogeneous, even though it is classified as a single phase. Supercritical region contains multiple structural regimes, which can be defined by different molecular ordering that is why liquid-like and gas-like terms are used widely. [12]

The physical interpretation is also strongly related to changes in temperature and pressure. When a system operates near the critical point even small changes in conditions can make huge variations in density and transport properties. This is especially important in flow systems like supercritical power cycles, where thermophysical properties can change during process. Studies have shown that these changes can have a significant impact on turbulence and heat transfer processes. [11]

3.2 Widom line and pseudo-boiling

Although there are no real phase transitions in supercritical regions, clear crossover regions can be detected that separate different regimes of fluid properties. One of the most widely used concepts to describe this is Widom line. It is defined by the maximum values of thermodynamic response functions, such as heat capacity and compressibility. Widom line represents the conditions where fluctuations in the fluid are strongest. [13]

Physically, the Widom line can be interpreted as a boundary between liquid-like and gas-like behavior in the supercritical region. On one side of the line fluid is denser and the interaction between molecules is stronger, while on the other side the fluid behaves more like a gas with lower density and weaker interactions. Even though the Widom line is not a real interface

between phases, it still offers a useful way to structure different regimes in supercritical region. [12]

Closely related to the Widom line is the concept of pseudo-boiling, which is one of the key characteristics of supercritical fluids. Pseudo-boiling means fast but continuous change in fluid properties, when temperature rises above pseudo-critical temperature in certain supercritical pressure. Unlike normal boiling, where there is a clear interface in phase transition, in pseudo-boiling there is no phase transition and no two-phase interface. [7] Instead, pseudo-boiling appears as a sharp but continuous variation in several thermophysical properties. For example, density can decrease rapidly with increasing temperature, while the specific heat capacity reaches a maximum at the pseudo-critical temperature. This behavior is associated with significant structural changes within the fluid dense, liquid-like regions break down and intermolecular distances increase, causing the fluid to transition toward gas-like behavior. However, this transformation occurs smoothly without any discontinuity. [14]

Pseudo-boiling can therefore be interpreted as a structural transition from liquid-like to gas-like behavior within the supercritical state. During this process, energy fluctuations in the fluid become very large, which explains the peak observed in heat capacity. The strong variation in density leads to pronounced property gradients that can significantly influence flow and heat transfer characteristics. Pseudo-boiling appears in the vicinity of the Widom line. The Widom line shows the thermodynamic conditions, where the structural transition associated with pseudo-boiling is most intense. [15]

The importance of pseudo-boiling is particularly evident in high-pressure and trans-critical flows. The sharp gradients in density and heat capacity near the pseudo-critical region can shape turbulence structures, affect boundary layer development, and either improve or weaken heat transfer. That is why pseudo-boiling plays a crucial role in the analysis and design of systems operating under supercritical conditions. Pseudo-boiling describes a rapid structural transformation in supercritical fluid. Pseudo-boiling is term for liquid-like fluid transfer to gas-like or vice versa, without a real phase change. It provides an essential framework for understanding how fluid properties can vary over a narrow temperature range, even though the system remains in a single phase. [11]

3.3 Comparison with Liquid and Gas Phases

Supercritical fluids have properties that are in between liquids and gases, making them very different from either normal fluid phase. Liquids are characterized by high density and strong intermolecular interactions, and gases have low density and weak interactions. Supercritical fluids combine features of both depending on thermodynamic conditions and which substance is in processing. This hybrid behavior is result from the absence of a clear phase boundary and the continuous variation of properties above the critical point. [10]

Density of supercritical fluids can have liquid-like values, especially at pressures well above the critical pressure. At the same time molecular mobility remains significantly higher than in liquids and reminds more of gas-like behavior. This results in relatively high diffusivity. Such a combination allows supercritical fluids to penetrate porous materials more effectively than liquids while maintaining stronger solubility than gases. [16]

Liquids are typically characterized by short-range order, while gases have not significant molecular organization. Supercritical fluids can display intermediate behavior, where transient clusters and local ordering appear and disappear dynamically. This leads to a continuously evolving microstructure that depends strongly on temperature and pressure. Crossing regions such as the Widom line represents conditions, where this microstructure change is the most intense. [14]

Thermodynamic response functions like Widom line highlight the unique position of supercritical fluids between liquid and gas phases. For example, near the pseudo-critical region, properties such as heat capacity and compressibility can reach high peaks, which are not seen in normal liquid or gas states. These anomalies are associated with strong density fluctuations and structural rearrangements within the fluid, reflecting its transitional nature. [15]

4 Thermophysical properties in supercritical state

4.1 Density and Compressibility

Density and compressibility are among the most important thermophysical properties governing the behavior of supercritical fluids. Unlike in conventional liquid or gas phases, these properties exhibit strong and continuous variation in the supercritical region, particularly near the critical point and along the pseudo-critical line. This sensitivity is a direct consequence of the absence of a phase boundary and the presence of large density fluctuations in the fluid. [10]

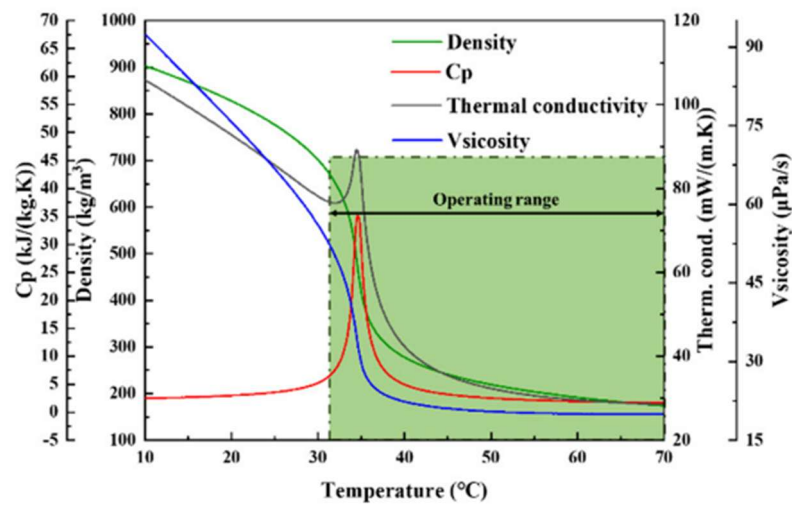


Figure 4: Thermodynamic properties of supercritical CO₂ in 8 MPa. [17]

In the supercritical state, density can vary over a wide range depending on pressure and temperature, as shown in Figure 4. At pressures slightly above the critical pressure, the fluid may have gas-like densities at high temperatures and liquid-like densities at lower temperatures. This change is continuous, meaning that no true phase change occurs. Nevertheless, near the pseudo-critical region, thermophysical properties can vary sharply with temperature and pressure, which strongly affects heat-transfer behavior. Near the pseudo-critical temperature, density exhibits particularly steep gradients with respect to temperature. [16]

Even a small increase in temperature can lead to a rapid decrease in density, especially in the pseudo-boiling region. This behavior is associated with structural changes in the fluid, where liquid-like clusters break down and the system transitions toward a more gas-like state. These

rapid variations in density play a critical role in determining flow behavior and heat transfer characteristics in supercritical systems. [17]

Isothermal compressibility describes how the volume of fluid responds to changes in pressure while temperature remains constant. In the supercritical region, especially near the critical point, compressibility can increase significantly due to large density fluctuations. This means that even small changes in pressure can result in large changes in density. This kind of behavior is not seen in normal liquid or gas states, where compressibility remains relatively low in liquids and constant in gases. [10]

In regions close to the coexistence line or pseudo-critical conditions, peaks in compressibility often coincide with rapid density changes. These phenomena are linked to the underlying molecular structure of the fluid and reflect the transition between liquid-like and gas-like behavior. As a result, accurate prediction of these properties is essential for modeling and designing systems operating under supercritical conditions. [18]

4.2 Specific Heat Capacity

Specific heat capacity is one of the most significant thermophysical properties of supercritical fluids, particularly because of their highly non-linear behavior near the critical and pseudo-critical regions. In general, the isobaric specific heat capacity describes the amount of heat required to raise the temperature of a substance at constant pressure. In supercritical fluids, this property shows behavior that is markedly different from that observed in conventional liquid or gas phases.

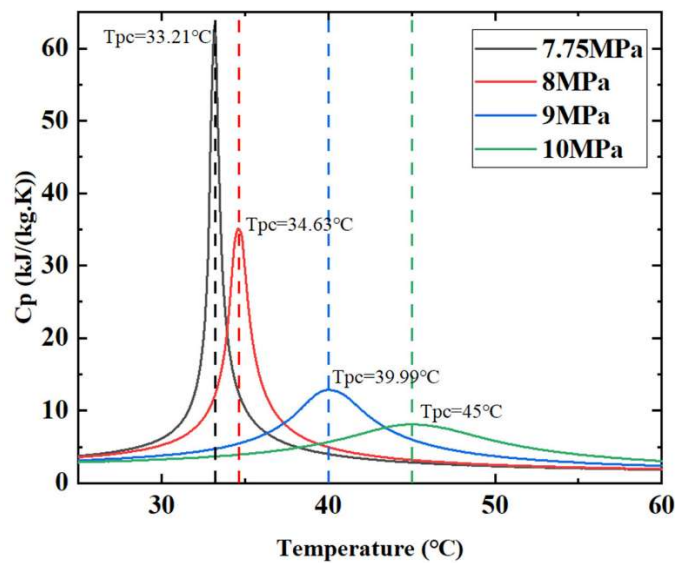


Figure 5: Specific heat capacity of supercritical CO₂ at different pressures. [17]

Far from the critical point, the specific heat capacity of a supercritical fluid behaves similarly to that of either a compressed liquid or a gas, depending on the density. However, as the system approaches the pseudo-critical temperature at a given pressure, specific heat increases sharply and typically reaches a pronounced maximum, as shown in Figure 5. [17] Near the pseudo-critical region, the fluid has a continuous transition from liquid-like to gas-like, often referred to as pseudo-boiling. During pseudo-boiling, a significant amount of energy is absorbed by the system without a corresponding large increase in temperature. Usually, temperature rise increases kinetic energy of molecules, but in supercritical region the supplied heat breaks intermolecular interactions and reorganizes the fluid structure. This results in as a peak in the specific heat capacity. [19]

The position and magnitude of the specific heat capacity peak depend strongly on pressure. When pressure increases above the critical value, the pseudo-critical temperature shifts, and the peak becomes broader and not so high as seen in Figure 5. This indicates that the sharpness of the pseudo-boiling process decreases at higher pressures. [15]

4.3 Viscosity

Viscosity is one of the most important transport properties which tells the resistance of fluids to flow. In supercritical fluids, viscosity has very much different behavior than in normal liquid and gas phases. While liquids typically have high viscosity due to strong intermolecular interactions and gases have low viscosity due to weak interactions and large intermolecular distances. Supercritical fluids display intermediate behavior of both that depends strongly on thermodynamic conditions. As shown in Figure 4, viscosity keeps going down while density has its peak. This gives good example of intermediate behavior. [10]

Usually, viscosity increases with density. This means that at higher pressures where the fluid becomes more liquid-like the viscosity also increases. However, the viscosity of supercritical fluids usually remains lower than that of liquids and closer to gas-like values, as in Figure 4. This is because molecular mobility in the supercritical state is still relatively high, allowing for easier flow compared to normal liquid phases. So, supercritical fluids combine low viscosity with high density, which is useful for transport processes. [16]

Viscosity is in important role in determining flow characteristics, pressure drop and heat transfer in supercritical systems. Low viscosity improves flow and reduces energy losses, while

its sensitivity to thermodynamic conditions must be considered in system design. In high pressure flows, even small changes in viscosity, can have significant effect on turbulence and boundary layer behavior. [20]

4.4 Thermal conductivity

As seen in Figure 4 there is fourth thermophysical property called thermal conductivity. That is also important property that describes the ability of fluid to conduct heat. Thermal conductivity has different behaviors due to the strong interaction between molecular motion, density and thermodynamic fluctuations near the critical point. Compared to normal subcritical fluids, supercritical fluids often have improved heat transfer characteristics, especially in the vicinity of the pseudo-critical region. [17]

Thermal conductivity increases with density because closer intermolecular spacing allows energy to be transferred more efficiently from molecule to molecule. Therefore, supercritical fluids may have thermal conductivity values more like liquids. This is why supercritical fluids are favorable in heat transfer applications, where efficient thermal transport is the main thing. [16]

5 Engineering applications

5.1 Supercritical Power Cycles

Power cycles are one of the most interesting applications for supercritical fluids. This means that these cycles operate above the critical point of the working fluid. One of the most used fluid is carbon dioxide, which allows higher thermal efficiencies compared to conventional Rankine cycles. The supercritical carbon dioxide Brayton cycle has gained significant attention due to its compact design, high efficiency and ability to operate at moderate temperatures for example 500–700 °C. [1]

The advantage of supercritical power cycles lies in the elimination of phase change during compression and expansion processes. Because the working fluid remains in a single phase, compression work is significantly reduced compared to traditional steam cycles. The high density of supercritical fluids reduces the size of turbine machinery, enabling more efficient system designs. [17]

High density and favorable heat transfer characteristics improve the performance of these cycles. However, the strong variation of properties near the pseudo-critical region requires careful thermal and fluid dynamic design to ensure stable operation. Overall, supercritical power cycles represent a promising technology for future energy systems, including nuclear, solar and waste heat recovery applications. [17]

5.2 Other Industrial Applications

Supercritical fluids are widely used across various industrial applications. One of the most established applications is supercritical fluid extraction, where supercritical CO₂ is used as a solvent to extract valuable compounds from natural materials. This technique is commonly applied in the food, pharmaceutical and cosmetic industries because it is environmentally friendly, non-toxic and leaves no solvent residues. [16]

Supercritical fluids are also used in chemical processing and environmental technologies. For example, supercritical water oxidation enables efficient destruction of hazardous organic waste. Supercritical fluids are also utilized in material processing, including nanoparticle synthesis and coating technologies, where precise control of density and solubility allows shaping material properties. [16]

Another important application is enhanced oil recovery. In this process supercritical CO₂ is injected into reservoirs to improve hydrocarbon extraction. Due to its low viscosity and high diffusivity, supercritical CO₂ can penetrate porous media effectively, while its relatively high density enhances dissolution of substances. These were just few applications what supercritical fluids have. But it is believed that supercritical fluids will have more useful applications in future, while the precision of older ones is growing. [21]

5.3 Advantages and Design Challenges

Supercritical fluids have several advantages that make them attractive for engineering applications. One of the key benefits is the ability to change fluid properties continuously by adjusting temperature and pressure. This allows optimization of density, viscosity and others for specific processes. All these features combined provides more efficient heat transfer and process technologies. [16]

Of course, there is many environmental benefits like in Supercritical CO₂, for example. SCO₂ is non-toxic, non-flammable and readily available that makes it a sustainable alternative to conventional organic solvents. In power generation, supercritical cycles can achieve higher efficiencies and lower emissions compared to traditional technologies, which reduces environmental load obviously. [1]

Despite these advantages there are still significant design challenges. One of the most common issues is the strong variation of thermophysical properties near the critical and pseudo-critical region. Rapid and strong changes in density, specific heat capacity and other properties can lead to complex flow behavior. This makes accurate modeling and control very difficult. These effects can significantly influence heat transfer, pressure drop and system stability. [17]

6 Conclusions

The fundamental behavior and properties of supercritical fluids have been examined from both thermodynamic and physical perspectives. Unlike traditional phases of matter, supercritical fluids exist beyond the critical point where the clear interface between liquid and gas disappears. Supercritical fluids have complex and highly discontinuous behavior and can have very different values in their properties. This makes the supercritical state both scientifically interesting and practically important. It was shown that in the supercritical region, thermophysical properties undergo strong and continuous variations. These changes are closely related to large molecular level fluctuations, which significantly influence macroscopic behavior. Concepts such as the Widom line provide us with a useful framework for describing the gradual transition between liquid-like and gas-like regimes, but no true phase boundary exists.

It is also highlighted that supercritical fluid cannot be treated as completely homogeneous despite it is a single thermodynamic phase. Instead, they contain different structural and dynamic regimes that strongly depend on temperature and pressure. This complex behavior has a direct impact on thermophysical properties and those are critical in engineering applications. Supercritical fluids offer significant advantages due to their tunable properties. With supercritical fluids we can achieve more efficient energy systems and environmentally friendly processes. Technologies such as supercritical power cycles, extraction processes, and advanced chemical treatments demonstrate their wide industrial potential. However, the same property variations that make these fluids useful also create challenges in system design and control.

In conclusion, understanding the thermophysical properties and phase behavior of supercritical fluids is essential for both theoretical research and practical applications. Even though there is a lot of progress, the supercritical state remains an active area of study, because the math and modelling of transitions and properties is highly complicated, it needs plenty of computing power. Continued research is necessary, not only in fluid dynamics but in other fields of science also, to fully deploy the potential of supercritical fluids.

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