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Evaporation of a CO₂ Droplet in a High Temperature, Supercritical Pressure Environment

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Received: 13 October 2025; Accepted: 15 December 2025; Published: 28 February 2026

ABSTRACT: This study presents a numerical investigation of the transient relaxation dynamics of a near-critical CO₂ droplet immersed in a warmer supercritical environment composed of the same fluid. Three thermodynamic regimes were analysed: quasi-critical ($T_r = 1.01$, $P_r = 1.01$), transitional ($T_r = 2.01$, $P_r = 1.01$), and deep supercritical ($T_r = 5.01$, $P_r = 3.01$). The evolution of density, temperature, and velocity fields was examined to characterize the internal structure and stability of the interfacial transition layer. The evolution of density, temperature, and velocity fields highlights the competition between thermal diffusion, compressibility, and mass confinement in shaping the stability of the interfacial transition layer. Near the critical point, strong gradients and flux discontinuities emerge, consistent with known instabilities, whereas higher reduced conditions promote homogenization and stabilized transport. In the deep supercritical regime, smooth and nearly uniform fields indicate robust thermal stability. The model is validated against prior studies on droplet evaporation under supercritical and trans-critical conditions. Beyond theoretical insights, the results underline practical implications for advanced propulsion, heat transfer, and evaporation systems as well as for safe CO₂ supercritical storage and extraction processes in energy, aerospace, pharmaceutical, and materials industries.

KEYWORDS: Navier–Stokes equations; evaporation; supercritical pressure; temperature; density and velocity

1 Introduction

Droplet evaporation plays a central role in the performance of liquid-fuel combustion systems, including diesel engines, gas turbines, and rocket thrusters. In these systems, fuel is injected into the combustion chamber as a cloud of droplets, where it vaporizes and oxidizes to release thermal energy [1,2]. When temperature and pressure exceed the fluid's critical point, a supercritical environment emerges, fundamentally altering thermodynamic properties and eliminating the distinct liquid gas interface [3].

Under such conditions, evaporation mechanisms become unconventional and difficult to model. The nonlinear coupling of mass, momentum, and energy transport, combined with the absence of a sharp interface, challenges the validity of classical approaches. Since the 1950s, several models have been proposed including the well-known d²-law of Spalding [4] and Godsave [5], which assumes a linear decrease of droplet diameter squared with time. However, this law, even when extended to supercritical regimes, relies on assumptions that break down at elevated pressures [6–8].

Experimental studies have further revealed phenomena that classical models cannot fully explain, such as thermal instabilities, pseudo-boiling phenomena, and discontinuities in flow fields, often linked to

incomplete combustion and increased pollutant emissions [9–11]. Spalding [6] proposed that under critical conditions, the droplet behaves as a point source of fluid. Sánchez-Tarifa et al. [12] introduced a three-zone model (cold droplet, transition layer, quasi-static outer region), enabling estimation of the recession rate of the intermediate layer via temperature profile analysis. These studies confirmed the partial applicability of the d^2 -law in supercritical regimes, while highlighting its limitations near the critical point [13–15].

In response to these challenges, recent research has advanced the modelling of transcritical and supercritical evaporation. Ly et al. [16] identified four distinct regimes governing the transition from subcritical evaporation to supercritical vaporization and dense fluid–gas mixing, using a diffuse interface method coupled with molecular simulations. Cheng and Xia [17] emphasized the potential of supercritical CO₂ as a thermal transport fluid in advanced energy systems, citing its high thermal conductivity, diffusivity, and low viscosity. Despite these advances, several studies have shown that instability and experimental difficulties persist. Min et al. [18] reported non-monotonic profiles and transient instabilities in near-critical regimes, while Kang et al. [19] emphasized the efficient heat transfer characteristics of deep supercritical conditions. Majumdar et al. [20] observed ultrafast molecular reorganization, confirming the singularity of supercritical regimes. Simeoni et al. [21], and Lopes et al. [22] further underlined the experimental challenges in characterizing thermal instabilities and pseudo-boiling phenomena. In addition to these studies, recent reviews have broadened the context of advanced fluids and supercritical CO₂ applications. Furthermore, Awais et al. [23] reviewed nanofluid heat transfer and pressure drop performance, underlining the trade-off between enhanced thermal conductivity and increased flow resistance. While nanofluids rely on particle-driven mechanisms, the present study focuses on pure CO₂ droplets, thereby contributing complementary insights into high-performance thermal systems. These results point to the need for a unified framework capable of bridging unstable near-critical dynamics with stabilized deep supercritical behavior, while distinguishing physical instabilities from numerical artifacts.

Taken together, these findings underline that important gaps remain in our understanding of droplet evaporation under supercritical conditions. Specifically: (i) the lack of quantitative validation of numerical models against experimental data near the critical point, (ii) the difficulty of distinguishing physical instabilities from numerical artifacts, and (iii) the absence of a unified framework capable of capturing transient relaxation dynamics in supercritical regimes. These bottlenecks limit predictive capability and hinder the design of efficient propulsion and energy systems.

To address these gaps, the present study introduces a multi-order perturbation framework based on the compressible Navier Stokes equations and a Van der Waals thermodynamic formulation. This approach captures non-ideal fluid behaviour, including molecular interactions and density fluctuations near the critical point. CO₂ is selected as the working fluid due to its chemical stability, low viscosity, and favourable critical coordinates ($T_c = 31.1^\circ\text{C}$, $P_c = 73.8\text{ bar}$) [24]. The objective is to analyse the transient relaxation dynamics of a cold CO₂ droplet immersed in a warmer supercritical environment, focusing on the structure and evolution of the interfacial transition layer and the spatiotemporal behavior of velocity, density, and temperature fields. A numerical approach is implemented, solving the conservation equations with appropriate discretization and parametric control of evaporation conditions. By explicitly addressing the limitations of classical models and experimental bottlenecks, this work aims to provide new insights into the mechanisms governing droplet evaporation in supercritical regimes.

2 Mathematical Model

We consider a cold droplet of pure carbon dioxide (CO₂) suddenly introduced into an infinite hotter supercritical environment composed of the same fluid. The droplet is initially at rest, with uniform temperature *equal to the critical temperature* $T_c(\text{K})$, density *equal to the critical density* $\rho_c(\text{Kg/m}^3)$, and

radius R_{in} (m). The surrounding medium is also quiescent, characterized by temperature T_{∞} (K), pressure P_{∞} (Pa), and density ρ_{∞} (Kg/m³). In the absence of external body forces and forced convection, the system preserves spherical symmetry over time.

Let r' denote the radial distance from the droplet center and $R'(t)$ its instantaneous radius. The flow velocity is denoted by $u'(r', t')$, where t' is time. The thermodynamic behavior is governed by a non-ideal Van der Waals equation of state, which relates local pressure $P'(r', t')$ to density $\rho'(r', t')$ and temperature $T'(r', t')$.

The following assumptions are adopted to simplify the physical model:

- The fluid is pure carbon dioxide (CO₂), treated as a single component fluid.
- The droplet is perfectly spherical and isolated, allowing the problem to be reduced to a radial geometry in spherical coordinates.
- Viscosity, gravity, and surface tension are neglected; pressure is assumed spatially uniform.
- The regime is supercritical: $T > T_c = 31.1^{\circ}\text{C}$; $P > P_c = 73.8$ bar.
- Evaporation is driven purely by thermal gradients; vapor diffusion is neglected.
- The thermodynamic behavior is modeled using a realistic Van der Waals equation of state, accounting for molecular interactions and excluded volume effects.
- The heat capacity at constant volume, C_v is assumed invariant and equal to that of an ideal gas.

Under these assumptions, the governing equations reduce to the spherically symmetric inviscid conservation laws for mass, momentum, and energy, expressed as follows [13,15]:

$$\begin{cases} \frac{\partial \rho'}{\partial t'} + \nabla \cdot (\rho' \vec{u}') = 0 \\ \rho' \left(\frac{\partial \vec{u}'}{\partial t'} + \vec{u}' \cdot \nabla \vec{u}' \right) = -\nabla P' \\ \rho' C_v \left(\frac{\partial T'}{\partial t'} + \vec{u}' \cdot \nabla T' \right) = \nabla \cdot (\lambda \nabla T') - (P' + a\rho'^2) \nabla \cdot \vec{u}' \\ P' = \frac{\rho' R T'}{1 - b'\rho'} - a\rho'^2 \end{cases} \quad (1)$$

The parameters a and b are determined from the experimental critical coordinates of the fluid under consideration. Specifically, the Van der Waals relations yield: $T_c = 8a/(27bR)$ and $\rho_c = 1/3b$. R denotes the universal gas constant.

These relations ensure that the non-ideal equation of state reproduces the correct critical point of CO₂. Based on these definitions, the governing conservation equations are subsequently reduced to a one-dimensional (1D) dimensionless form under spherical symmetry [13]:

$$\begin{cases} \frac{\partial \rho'}{\partial t'} + \frac{1}{r'^2} \frac{\partial (r'^2 \rho' u')}{\partial r'} = 0 \\ \rho' \left(\frac{\partial u'}{\partial t'} + u' \frac{\partial u'}{\partial r'} \right) = -\frac{\partial P'}{\partial r'} \\ \rho' C_v \left(\frac{\partial T'}{\partial t'} + u' \frac{\partial T'}{\partial r'} \right) = \frac{1}{r'^2} \frac{\partial}{\partial r'} \left(r'^2 \lambda' \frac{\partial T'}{\partial r'} \right) - (P' + a'\rho'^2) \left(\frac{2u'}{r'} + \frac{\partial u'}{\partial r'} \right) \\ P' = \frac{\rho' R' T'}{1 - b'\rho'} - a'\rho'^2 \end{cases} \quad (2)$$

In the critical and supercritical regimes, the thermal conductivity of CO₂ is expressed as follows [25]:

$$\lambda'(\rho') = \lambda'_o \left(\sqrt{T'/T_{\infty}} + 0.078 * \frac{\rho'}{\rho_c} + 0.014 * \left| \frac{T'}{T_c} - 1 \right|^{-1/3} \right) \quad (3)$$

Eq. (3) expresses effective thermal conductivity as a function of reduced temperature and density. The first term $\sqrt{T'/T'_\infty}$ represents classical thermal diffusion, the second term $0.078 \cdot \rho'/\rho'_c$ accounts for molecular cohesion through density dependence, and the third term $0.014 \cdot |T'/T'_c - 1|^{-1/3}$ captures critical corrections near T'_c . This last contribution becomes dominant in the near-critical regime, explaining the unstable oscillations and sharp gradients. Away from the critical point, diffusion and density terms prevail, yielding smoother relaxation profiles. Thus, Eq. (3) provides a direct link between mathematical formulation and the physical mechanisms underlying the simulated temperature fields.

A constant temperature T'_∞ is imposed at the external shell boundary, whereas a zero-heat flux condition is enforced at the droplet center ($r' = 0$). The governing variables are nondimensionalized with respect to the critical properties of the CO_2 using the following dimensionless quantities:

$$\rho = \frac{\rho'}{\rho'_c}; T = \frac{T'}{T'_c}; P = \frac{P'}{\rho'_c R' T'_c}; u = \frac{u'}{C'_o} \text{ and } t = \frac{t'}{\frac{R'_{in}}{V'}}, r = \frac{r'}{R'_{in}}$$

where V' is the characteristic velocity and C'_o the speed of sound in a perfect gas at T'_c . Under low Mach number ($Ma = \frac{V'}{C'_o}$), compressibility effects can be negligible and the flow can be treated as quasi-incompressible. Two characteristic times are introduced: t_a , which is the reference acoustic time in a perfect gas at T'_c , and t_{diff} , which is the thermal diffusion time in the same gas. By substituting the dimensionless variables into the governing system presented in Eq. (2) and performing an asymptotic expansion with respect to the Mach number Ma [26], the first-order and second-order approximation systems in Eqs. (4) and (5), are obtained.

First approximation system Eq. (4):

$$\begin{cases} \frac{\partial \rho_0}{\partial t} + \frac{1}{r^2} \frac{\partial [r^2 \rho_0 U_0]}{\partial r} = 0 \\ \rho_0 \left(\frac{\partial U_0}{\partial t} + U_0 \frac{\partial U_0}{\partial r} \right) = -\frac{1}{\gamma} \frac{\partial P_0}{\partial r} \\ P_0 = \frac{\rho_0 T_0}{1-b\rho_0} - a\rho_0^2 \\ \rho_0 \left(\frac{\partial T_0}{\partial t} + U_0 \frac{\partial T_0}{\partial r} \right) = \frac{t_a}{t_{diff}} \frac{\gamma}{Ma} \frac{1}{r^2} \frac{\partial [r^2 \lambda \frac{\partial T_0}{\partial r}]}{\partial r} - (\gamma-1) \frac{\rho_0 T_0}{1-b\rho_0} \left(\frac{2U_0}{r} + \frac{\partial U_0}{\partial r} \right) \end{cases} \quad (4)$$

Second approximation system Eq. (5):

$$\begin{cases} \frac{\partial \rho_1}{\partial t} + \frac{1}{r^2} \frac{\partial [r^2 (\rho_1 U_0 + \rho_0 U_1)]}{\partial r} = 0 \\ \rho_0 \left(\frac{\partial U_1}{\partial t} + U_0 \frac{\partial U_1}{\partial r} + U_1 \frac{\partial U_0}{\partial r} \right) + \rho_1 \left(\frac{\partial U_0}{\partial t} + U_0 \frac{\partial U_0}{\partial r} \right) = -\frac{1}{\gamma} \frac{\partial P_1}{\partial r} \\ P_1 = \frac{\rho_1 T_0 + \rho_0 T_1}{1-b\rho_0} - a\rho_0 \rho_1 \\ \rho_0 \left(\frac{\partial T_1}{\partial t} + U_0 \frac{\partial T_1}{\partial r} + U_1 \frac{\partial T_0}{\partial r} \right) + \rho_1 \left(\frac{\partial T_0}{\partial t} + U_0 \frac{\partial T_0}{\partial r} \right) = \frac{t_a}{t_{diff}} \frac{\gamma}{Ma} \frac{1}{r^2} \frac{\partial [r^2 \lambda \frac{\partial T_1}{\partial r}]}{\partial r} - (\gamma-1) \frac{(\rho_1 T_0 + \rho_0 T_1)}{1-b\rho_0} \left(\frac{2U_0}{r} + \frac{\partial U_0}{\partial r} \right) \end{cases} \quad (5)$$

where:

- ρ_0, ρ_1 are zeroth- and first-order density perturbations
- U_0, U_1 are zeroth- and first-order velocity perturbations
- T_0, T_1 are zeroth- and first-order temperature perturbations
- P_0, P_1 are zeroth- and first-order pressure perturbations

with the initial conditions:

$$T(0, r) = \begin{cases} T_c, & r \leq R_{in} \\ T_\infty(r), & r > R_{in} \end{cases}$$

$$\rho(0, r) = \begin{cases} \rho_c, & r \leq R_{in} \\ \rho_\infty(r), & r > 0 \end{cases} \quad (6)$$

the boundary conditions:

$$\begin{aligned} T(t, r) &= T_c, r \leq R_{in} \\ \lim_{r \rightarrow +\infty} T(r, t) &= T_\infty \\ \rho(t, r) &= \rho_c, r \leq R_{in} \\ \lim_{r \rightarrow +\infty} \rho(r, t) &= \rho_\infty \end{aligned} \quad (7)$$

A constant wall-temperature boundary condition was imposed to isolate the intrinsic effects of thermodynamic regimes, without introducing additional convective coupling. This choice highlights the fundamental differences between thermal confinement, relaxation, and homogenization across the regimes studied. Consequently, the simulations were carried out using CO₂ as the working fluid, with temperature-dependent thermophysical properties near the critical point.

3 Numerical Resolution

The system is nondimensionalized using critical fluid properties, and an asymptotic expansion with respect to the Mach number yields zeroth- and first-order perturbation systems. Due to the nonlinearity of the governing equations, no analytical solution was possible, therefore, the equations were solved numerically via a coupled finite volume approach [27–29] with second-order centered spatial discretization and an Crank-Nicolson scheme ensuring stability at large time steps. The domain is discretized into concentric radial cells, refined near the droplet-fluid interface to capture steep thermal gradients, while convective terms were linearized using a Newton-Raphson method. A mesh independence study was conducted across three refinement levels, with the convergence criterion defined as:

$$\frac{\max |\rho_N - \rho_{N'}|}{\max |\rho_{N'}|} < \delta \quad (8)$$

where (N) and (N') are two successive meshes, and δ is the convergence threshold. A mesh of 150 cells was selected, as variations in the density and temperature profiles were below 1%, ensuring sufficient accuracy without excessive computational cost. Iterations continued until the residuals of conservation equations fell below 10^{-6} for the primary variables (ρ , T and u). The model was validated against the numerical results of Sagna and d'Almeida [3,13], addressing droplet evaporation under supercritical conditions. In addition the resulting temperature and velocity profiles were also compared with the numerical results of Ly et al. [16], concerning droplet evaporation in both supercritical and transcritical regimes.

4 Results and Discussion

The numerical simulations reveal distinct evaporation behaviors across the three thermodynamic regimes. In the quasi-critical regime, the droplet maintains a sharp thermal gradient, and the transition layer remains thin and well-defined. As the system enters the transitional regime, the interfacial layer broadens, and nonlinear coupling between temperature and velocity fields becomes pronounced, leading to nonmonotonic relaxation profiles and localized thermal waves. It is worth noting that, although the model relies on spherical symmetry, the observed instabilities suggest that, if this symmetry were broken, they would manifest anisotropically, leading to non-uniform evaporation rates and asymmetric redistribution of mass and heat.

4.1 Velocity Evolution in the Droplet

Figs. 1 and 2 illustrate the spatiotemporal evolution of the flow velocity within a carbon dioxide droplet immersed in a supercritical environment, evaluated at zeroth and first order, respectively. Both figures correspond to reduced temperature $T_r > 1$ and reduced pressure $P_r > 1$, representative of supercritical conditions.

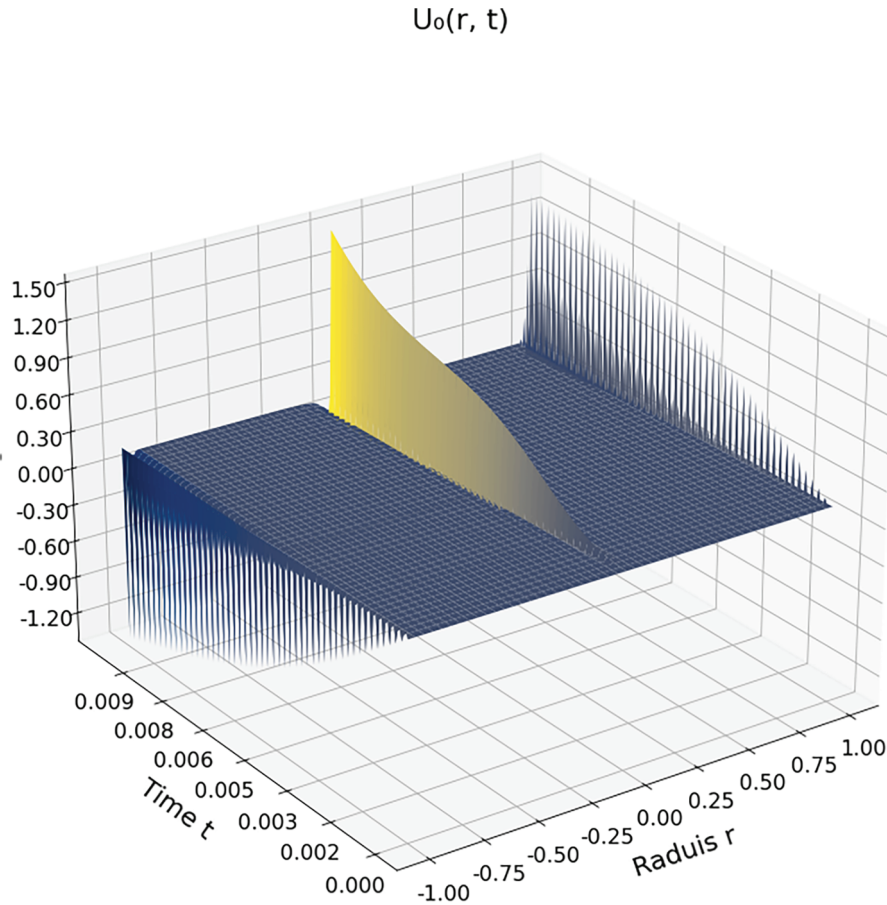


Figure 1: Zeroth-order velocity field $U_0(r, t)$ in a CO_2 droplet at $T_r = 1.01$, $P_r = 1.01$: emergence of internal extrema and compressibility effects

Fig. 1 shows the zeroth-order velocity field $U_0(r, t)$ which remains spatially continuous across the domain. However, its temporal evolution reveals nonmonotonic behavior, characterized by the emergence of localized extrema within the droplet. These features suggest the presence of internal perturbations propagating radially toward the interface. Although the zeroth-order regime is quasi-incompressible by construction, the observed dynamics reflect transient thermodynamic instabilities and early-stage compressibility effects triggered by the sudden exposure to supercritical conditions.

Fig. 2 presents the first-order velocity correction $U_1(r, t)$, which captures finer-scale variations and nonlinear responses. The field exhibits sharper gradients and pronounced peaks, indicating increased sensitivity to initial conditions and stronger coupling between thermodynamic variables. These localized structures may correspond to relaxation fronts or wave-like features, consistent with the formation of thermal or mass discontinuities in supercritical fluids.

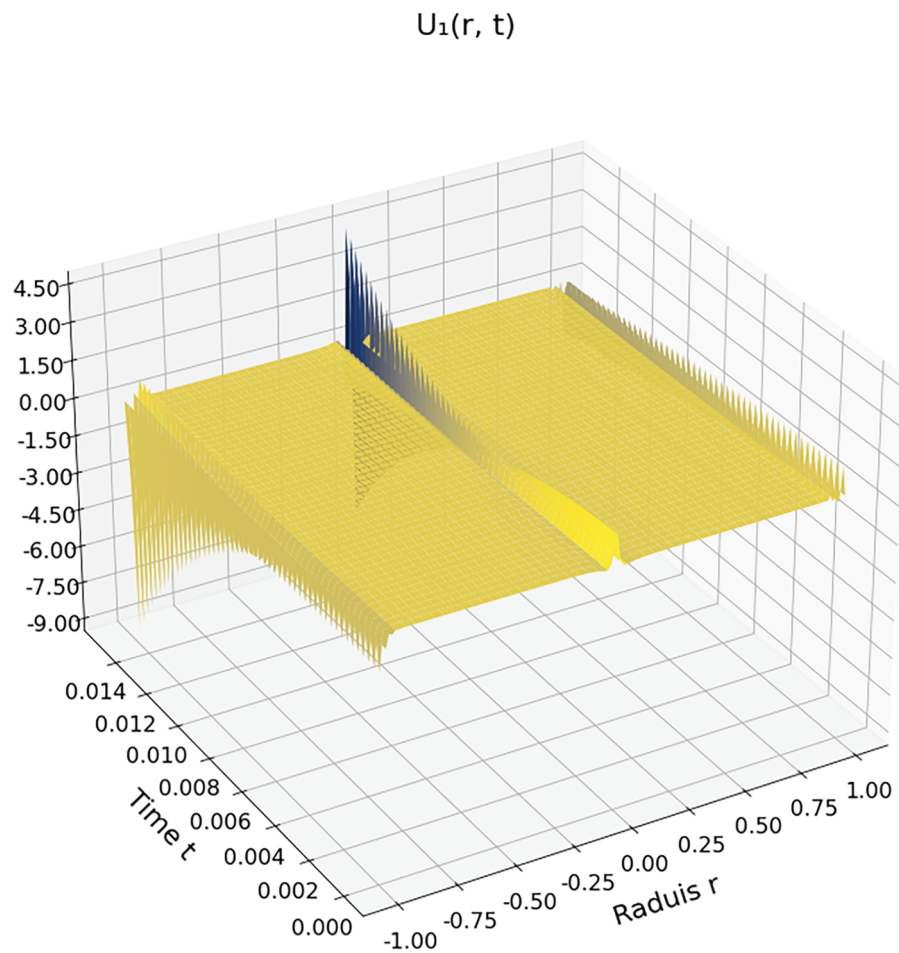


Figure 2: First-order velocity correction $U_1(r, t)$ at $T_r = 1.01$, $P_r = 1.01$: nonlinear gradients and shock-like structures

Fig. 3 compares radial velocity profiles $U_0(r)$ for two reduced temperatures ($T_r = 2.01$ and $T_r = 1.01$) and three values of reduced pressure ($P_r = 1.01, 2.02$ and 3.01). At constant temperature, increasing the reduced pressure P_r tends to stabilize the velocity field, producing more localized peaks and attenuated peripheral oscillations. This behavior reflects reduced compressibility and enhanced fluid cohesion. Conversely, at constant pressure, increasing T_r promotes thermal expansion, resulting in a smoother velocity field that is less responsive to localized perturbations.

Sharp velocity gradients often centred near $r = 0$, can be interpreted as signatures of hydrodynamic instabilities or relaxation fronts. These observations align with the results of Simeoni et al. [21], who emphasized the sensitivity of supercritical fluids to thermodynamic gradients and internal perturbations.

Thermodynamic instabilities manifest through non-monotonic velocity profiles and coherent relaxation fronts, in agreement with previous observations in supercritical fluids (e.g., Simeoni et al. [21]). In contrast, numerical instabilities are sensitive to discretization parameters such as time step and mesh resolution. Convergence and mesh-independence tests were performed, confirming that the oscillations and gradients reported here are robust and of physical origin.

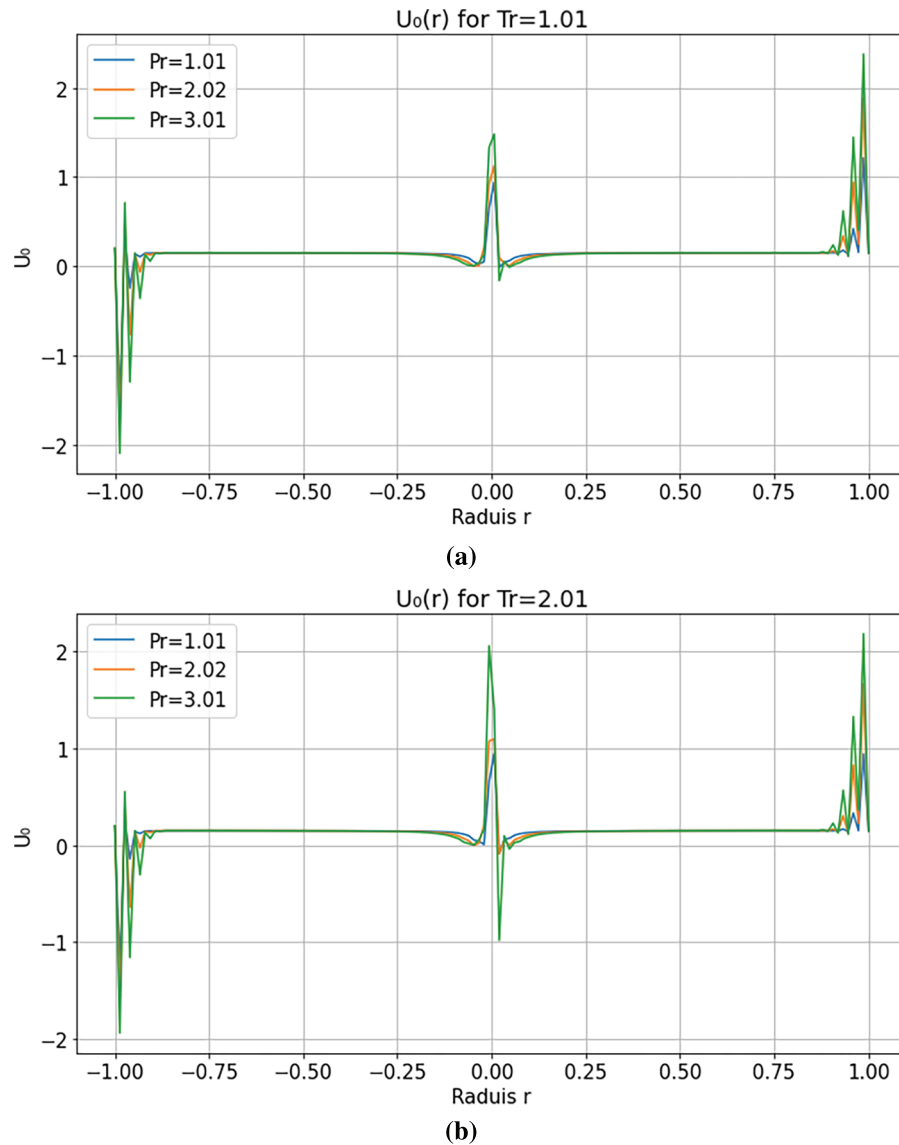


Figure 3: (a): Radial velocity profiles $U(r)$ for $T_r = 1.01$ comparison of compressibility-driven instabilities; (b): Radial velocity profiles $U(r)$ for $T_r = 2.01$: comparison of compressibility-driven instabilities

Overall, the radial velocity field emerges as a key indicator of transient regimes and coupled mechanisms involving thermal diffusion, mechanical compression, and interfacial instability. These results reinforce the relevance of multi-order modelling approaches for capturing the complex interplay between thermodynamic gradients, compressibility, and transient flow structures in supercritical droplet dynamics.

The quasi-critical regime exhibits localized perturbations and radial fronts propagating toward the interface. These structures can be interpreted as signatures of internal recirculation, contributing to non-monotonic velocity profiles and influencing the redistribution of mass and heat.

4.2 Density Evolution

The spatiotemporal evolution of the density field within a carbon dioxide droplet immersed in a supercritical environment reveals distinct dynamic regimes. Fig. 4 shows the zeroth-order density field

$\rho_0(r, t)$, characterized by the radial propagation of mass perturbations from the droplet center toward its periphery. These disturbances, while continuous, reflect transient compressibility effects and a progressive redistribution of mass triggered by the sudden exposure to supercritical conditions.

$$\rho_0(r, t)$$

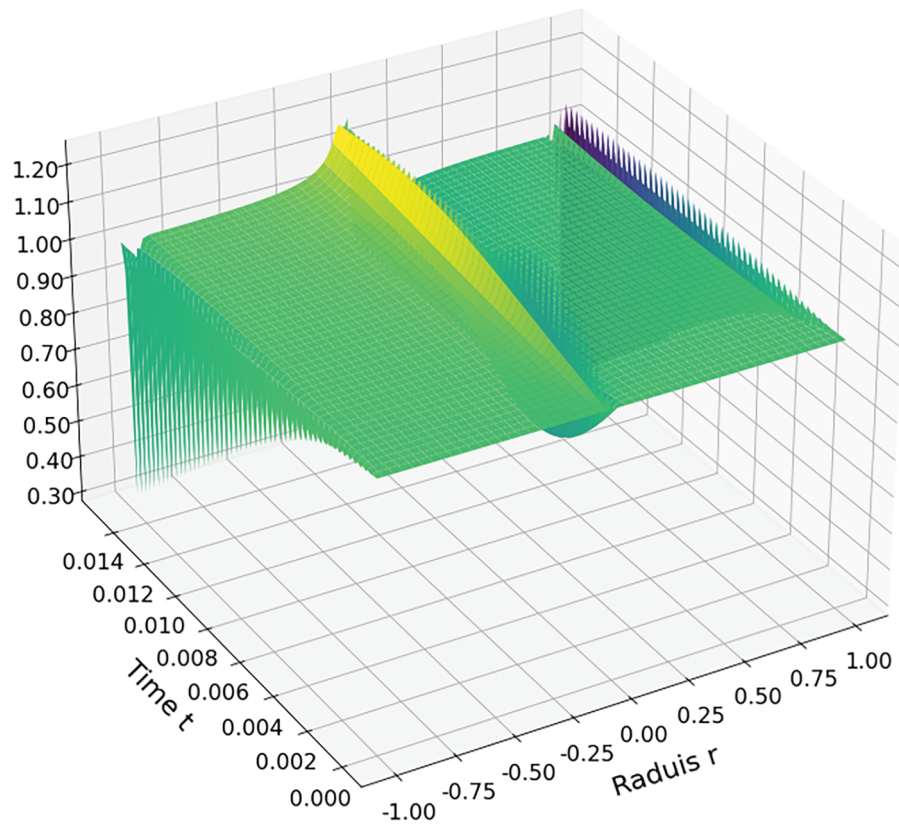


Figure 4: Zeroth-order density field $\rho_0(r, t)$ at $T_r = 1.01$, $P_r = 1.01$: radial propagation of mass perturbations

Fig. 5 presents the first-order correction $\rho_1(r, t)$, which captures nonlinear interactions and differential responses. Localized peaks and discontinuities emerge, suggesting the formation of relaxation fronts and density waves. These features indicate coupled mechanisms involving thermal diffusion, compressibility, and thermodynamic instabilities, particularly near the critical point.

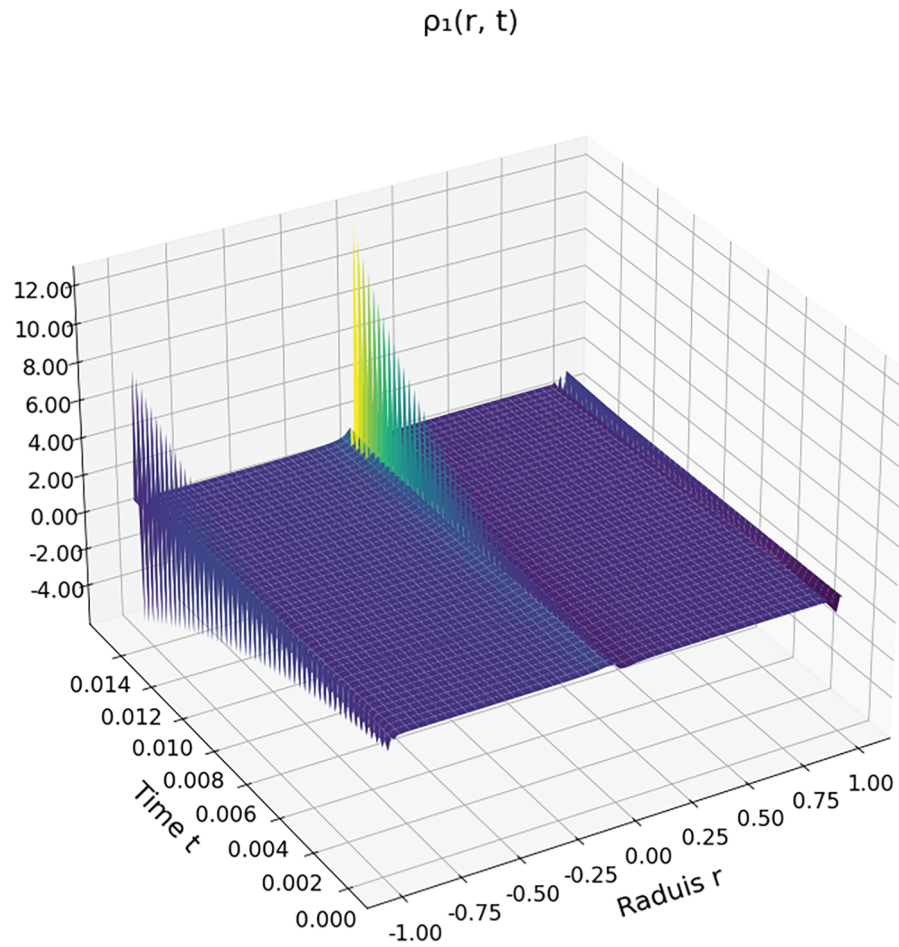


Figure 5: First-order density field $\rho_1(r, t)$ at $T_r = 1.01$, $P_r = 1.01$: emergence of relaxation fronts and nonlinear couplings

Figs. 6 and 7 illustrate the radial density profiles $\rho(r)$ for increasing reduced temperatures and pressures. At low $T_r = 1.01$, the profiles exhibit pronounced oscillations and peripheral instabilities, especially at low P_r , reflecting high compressibility and sensitivity to boundary conditions. As P_r increases, the central density peak intensifies and fluctuations diminish, indicating a transition toward a more confined and thermodynamically stable regime.

At moderate and high reduced temperatures ($T_r = 2.01$ and $T_r = 5.01$), the profiles become increasingly symmetric and sharply concentrated around $r = 0$. The amplitude of the central peak rises with P_r , suggesting enhanced mass confinement and reduced diffusivity at the droplet core. Peripheral attenuation at high temperature reflects a diffusion-dominated regime, where thermal expansion smooths out density gradients.

These results are consistent with previous studies [9,30], which reported nonmonotonic density profiles and interfacial instabilities in supercritical droplets. They underscore the relevance of multi-order modeling for capturing transient regimes and phase-transition dynamics in supercritical environments.

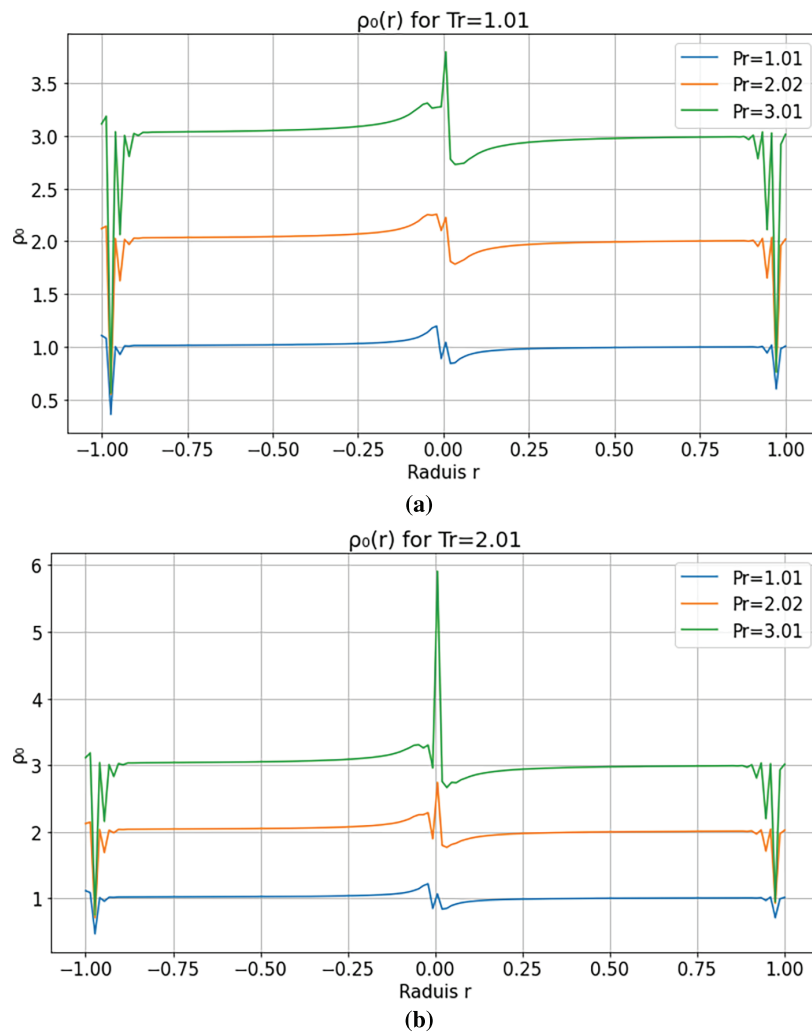


Figure 6: (a): Transition from unstable to confined regimes radial density profiles $\rho(r)$ for $T_r = 1.01$; (b): Transition from unstable to confined regimes radial density profiles $\rho(r)$ for $T_r = 2.01$

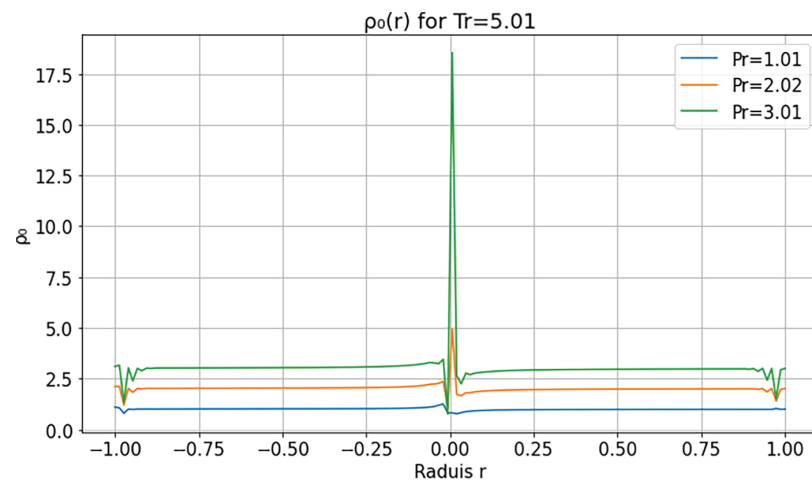


Figure 7: Radial density profile $\rho(r)$ at $T_r = 5.01$: deep supercritical confinement and mass concentration

The observed velocity and density gradients reflect a competition between thermal diffusion, compressibility, and mass confinement. At low reduced pressure/temperature, enhanced compressibility amplifies the system's sensitivity to perturbations, producing peripheral oscillations and radially propagating relaxation fronts. At higher reduced pressure/temperature, stronger molecular cohesion and more uniform thermal expansion damp local disturbances, yielding a confined and stable regime. Non-monotonic profiles therefore indicate an out-of-equilibrium response where diffusion does not instantaneously offset compressibility-induced gradients. These interpretations are consistent with experimental reports [2,31].

4.3 Evolution of the Temperature in the Droplet

Figs. 8–14 illustrate the temperature field evolution within a CO₂ droplet subjected to various supercritical regimes, governed by changes in reduced temperature and pressure.

4.3.1 Near-Critical Regime—Figs. 8 and 9 ($T_r = 1.01$, $P_r = 1.01$)

The zeroth-order temperature field $T_0(r)$ displays a narrow peak centered at $r = 0$, indicating localized thermal accumulation at the droplet core. This behavior is typical of near-critical conditions, where thermal diffusivity drops sharply, slowing energy dissipation and promoting intense thermal confinement. Lopes et al. [22] demonstrated that classical heat transfer correlations become inadequate in this regime, requiring specific models to describe the observed gradients.

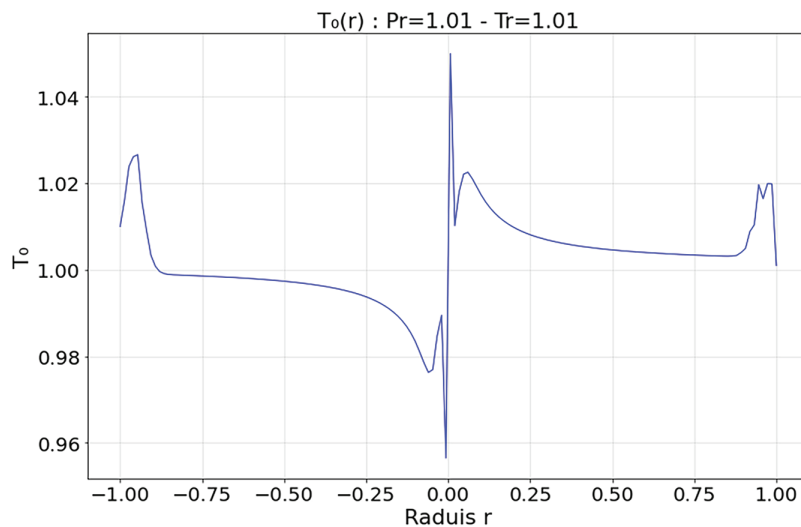


Figure 8: Spatial evolution zeroth-order heat flux within the droplet at $T_r = 1.01$ and $P_r = 1.01$

The first-order field $T_1(r)$ reveals an amplified and symmetric peak with weak lateral oscillations, reflecting nonlinear thermodynamic coupling and unstable gradients. Chai & Tassou [32] showed that proximity to the critical point induces local variations in density and heat capacity, which drive such instabilities. Min et al. [18] further linked flow instabilities in supercritical CO₂ systems to density and temperature fluctuations, identifying an unstable zone around $T_r \approx 1.01$, $P_r \approx 1.01$, consistent with our observations.

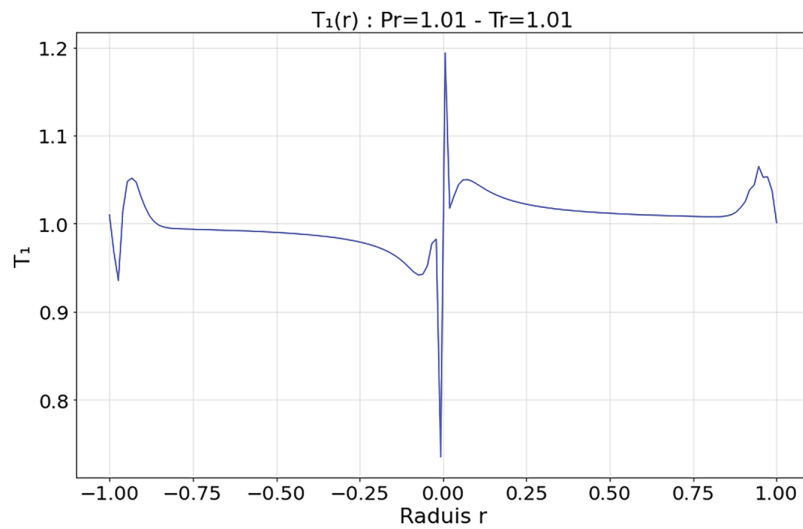


Figure 9: Spatial evolution first-order heat flux within the droplet at $T_r = 1.01$ and $P_r = 1.01$

4.3.2 Transitional Regime—Figs. 10 and 11 ($T_r = 2.01$, $P_r = 1.01$)

The zeroth-order field $T_0(r)$ becomes smoother, with a modest elevation near the center, suggesting moderate thermal concentration. The first-order field $T_1(r)$ shows a localized, symmetric peak with limited amplitude, indicating rapid damping of initial perturbations. This configuration reflects a thermodynamic regime dominated by controlled heat redistribution and pseudo-critical effects. The steep rise near the droplet boundary in $T_0(r)$, followed by a nearly uniform plateau, indicates localized thermal accumulation without global instability, while the sharp peak in $T_1(r)$ highlights strong thermal damping and relaxation dynamics. Min et al. [18] identified this regime as a thermal stability zone where density and temperature variations remain contained. Lopes et al. [22] noted that classical heat transfer models remain valid here, provided pseudo-critical effects are accounted for.

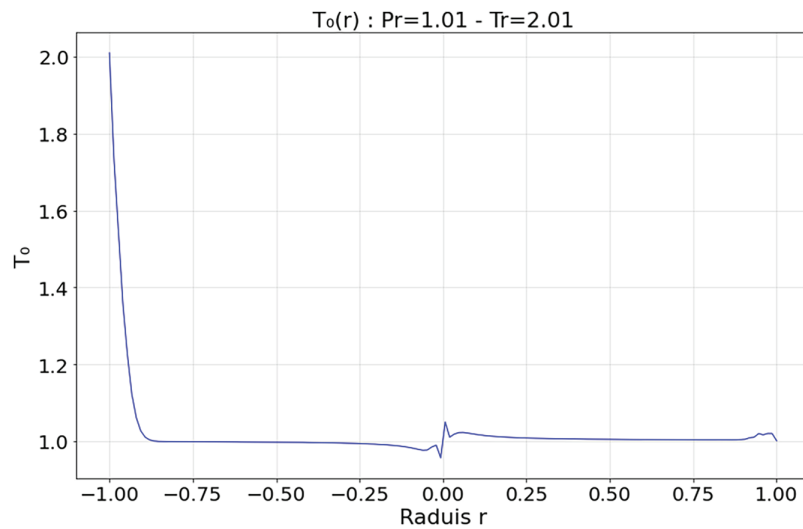


Figure 10: Spatial evolution zeroth-order heat flux within the droplet at $T_r = 2.01$ and $P_r = 1.01$

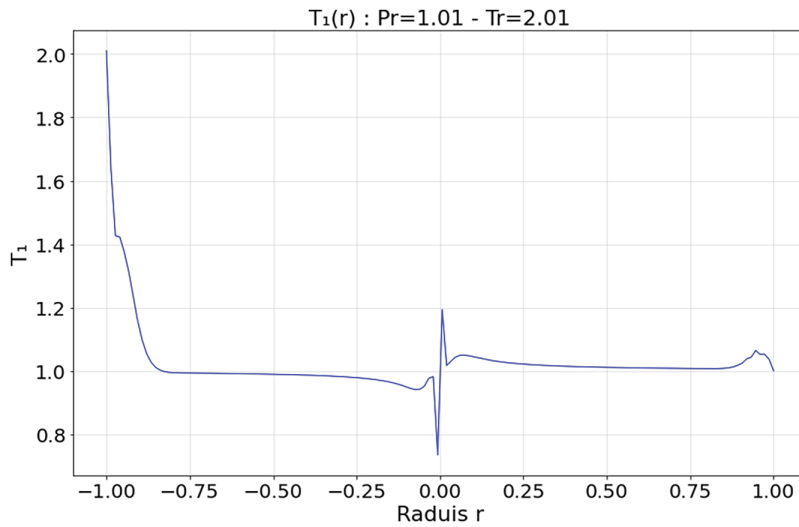


Figure 11: Spatial evolution first-order heat flux within the droplet at $T_r = 2.01$ and $P_r = 1.01$

Simeoni et al. [21] and Majumdar et al. [20] confirmed that this intermediate regime is marked by moderate thermal gradients and relaxation dynamics without phase rupture, characteristic of pseudo-boiling phenomena in supercritical CO_2 .

4.3.3 Deep Supercritical Regime—Figs. 12 and 13 ($T_r = 5.01$, $P_r = 3.01$)

The zeroth-order field $T_0(r)$ exhibits a nearly uniform profile, with slight central elevation and minimal boundary variations, indicating strong thermal homogeneity. The first-order field $T_1(r)$ shows a narrow central peak of moderate amplitude, without significant propagation or instability, reflecting ultrafast relaxation dynamics. This behaviour aligns with observations by Majumdar et al. [20] who reported, molecular clusters reorganization in deep supercritical states without generating macroscopic shock fronts.

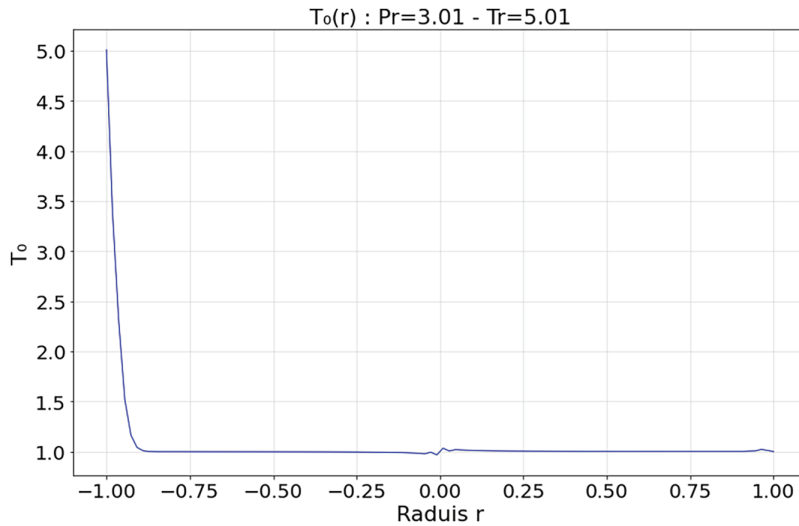


Figure 12: Spatial evolution zeroth-order heat flux within the droplet at $T_r = 5.01$ and $P_r = 3.01$

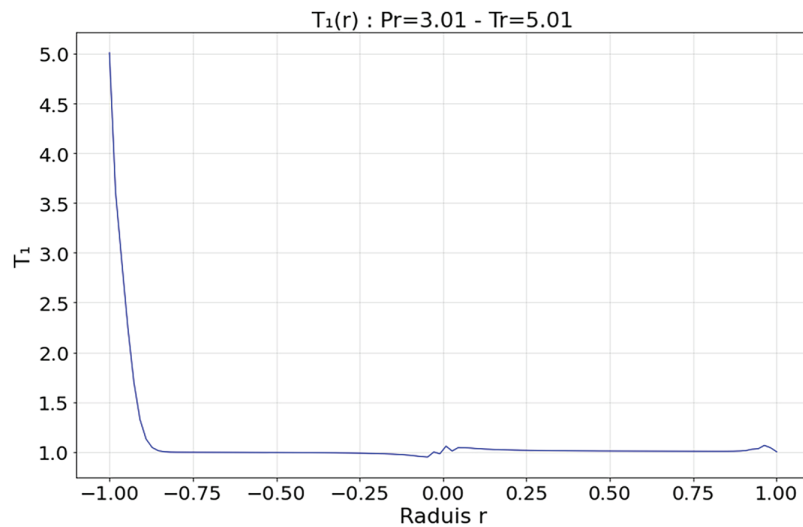


Figure 13: Spatial evolution first-order heat flux within the droplet at $T_r = 5.01$ and $P_r = 3.01$

The absence of a distinct interface between the droplet and the surrounding medium, limits the formation of shock fronts or transition zones or discontinuities, ensuring structural stability. Kang et al. [19] highlighted the efficiency and stability of deep supercritical regimes for heat exchange and solute dissolution, particularly in energy and space systems. Overall, this regime is characterized by low thermal gradients and homogeneous diffusion, confirming its suitability for high-performance applications under extreme conditions.

4.3.4 Radial Profiles—Figs. 14 and 15

At low reduced temperature ($T_r = 1.01$), the curves show localized oscillations around the center, indicating thermal instability induced by the imposed gradient between the droplet and the surrounding atmosphere (Fig. 14). At moderate temperature ($T_r = 2.01$), the profiles become smoother, with progressive thermal relaxation from the periphery toward the core (Fig. 15). The thermal peak near $r \approx -1$ corresponds to the initial contact zone with the hot atmosphere, while the central plateau indicates efficient thermal absorption without a distinct liquid-gas interface.

These results confirm that the choice of reduced parameters strongly influences the internal thermal dynamics of the droplet. Multi-order analysis enables accurate resolution of diffusion, relaxation, and stabilization mechanisms. Beyond regime-specific observations, these temperature fields highlight general mechanisms that govern supercritical droplet dynamics. Thermodynamic instabilities are identified through non-monotonic profiles and relaxation fronts, consistent with previous studies (e.g., Simeoni et al. [21]). In contrast, numerical instabilities depend on discretization parameters such as time step and mesh resolution. Convergence and mesh-independence tests were performed, confirming that the oscillations and gradients reported here are robust and of physical origin. Temperature profiles reveal the same competition between thermal diffusion, compressibility, and mass confinement observed in the velocity field. In the quasi-critical regime, the droplet sustains a sharp thermal gradient with early relaxation fronts, reflecting diffusion that lags the initial compressive transient. Entering the transitional regime, the interfacial layer broadens, and temperature becomes locally non-monotonic, with overshoot or plateau features indicative of nonlinear T-u-p coupling and thermally driven radial fronts. At higher reduced pressure/temperature, stronger cohesion and more uniform thermal expansion damp local disturbances, yielding smoother temperature profiles and

a less reactive interface. These trends support a coupled picture in which diffusion progressively dominates over compressibility, while internal recirculation can transiently recycle heat and sustain localized structures.

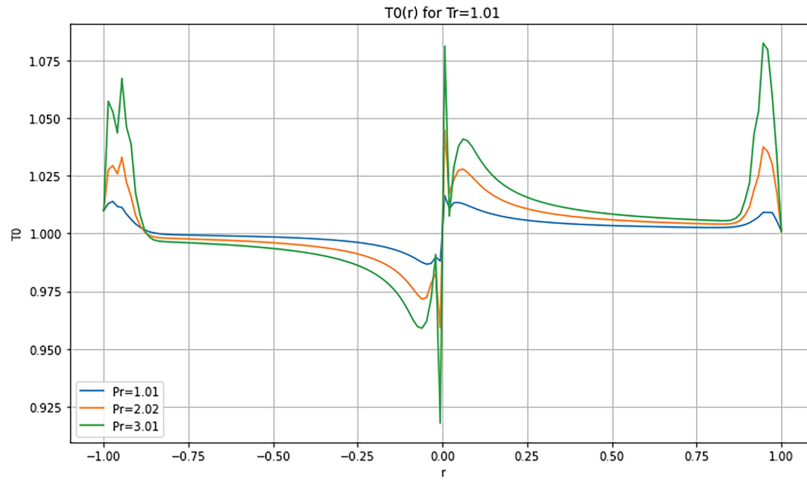


Figure 14: Radial Temperature distribution $T_0(r)$ for $Tr = 1.01$

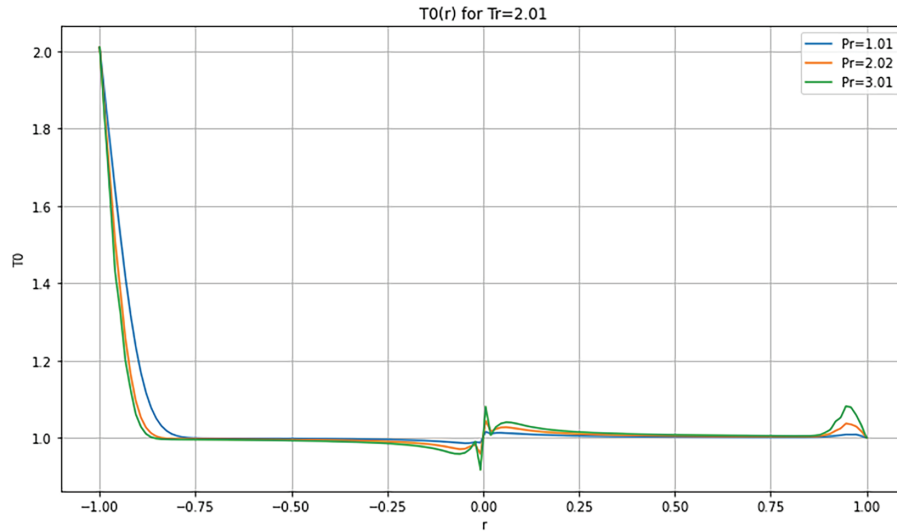


Figure 15: Radial Temperature distribution $T_0(r)$ for $Tr = 2.01$

It should be noted, however, that the present model relies on idealized assumptions, such as neglecting viscous losses, variations in heat transfer efficiency, and material aging. While these simplifications may limit direct applicability to real systems, they provide a tractable framework to highlight the dominant physical mechanisms. Future work will incorporate non-ideal effects to improve practical relevance.

5 Conclusion

Through detailed numerical simulations, this work characterizes the relaxation behaviour of a CO_2 droplet across three distinct thermodynamic regimes. In the quasi-critical case ($Tr = 1.01$, $Pr = 1.01$), sharp thermal gradients, localized density accumulation, and pronounced velocity perturbations are observed.

These features are consistent with flow instabilities and property fluctuations reported by Min et al. [18] and Lopes et al. [22].

In the transitional regime ($T_r = 2.01$, $P_r = 2.01$), thermal diffusion remains active but subdued, with moderate central heating and damped velocity oscillations. The system exhibits partial homogenization, bridging unstable near-critical dynamics and stabilized supercritical behavior. In the deep supercritical regime ($T_r = 5.01$, $P_r = 3.01$), the fields become nearly uniform, with minimal thermal or hydrodynamic disturbances. This regime reflects a stabilized transport behaviour, in line with ultrafast molecular reorganization observed by Majumdar et al. [20] and the efficient heat transfer characteristics described by Kang et al. [19].

The numerical framework based on second-order spatial discretization, Crank-Nicolson time integration, and mesh refinement near the interface proves robust and accurate. Validation against prior studies by Ly et al. [16] confirms the model's ability to reproduce key features of supercritical and transcritical droplet evaporation.

Overall, this study advances the understanding of transition layer dynamics in supercritical fluids and contributes to the development of predictive models for jet formation, transient evaporation, and high-performance thermal systems using CO₂ as a working fluid. The present results have direct industrial relevance. Regime-dependent thermal behaviors provide guidance for optimizing combustion and propulsion systems, where improved control of thermal instabilities can reduce fuel consumption and enhance energy efficiency. They also support the design of safe CO₂ supercritical storage and transport facilities, and the improvement of supercritical extraction processes in pharmaceutical, food, and materials industries. These perspectives underline that the proposed modeling approach offers practical insights that extend beyond theoretical analysis. Nevertheless, future work should address feasibility aspects such as energy demand and containment materials, as well as technical challenges including viscous dissipation, variable transport properties, and material aging, to strengthen the predictive capability and industrial applicability of the system.

Acknowledgement: Not applicable.

Funding Statement: The authors received no specific funding for this study.

Author Contributions: Conceptualization: Yendoubouame Lare, Koffi Sagna; methodology: Yendoubouame Lare; software: Yendoubouame Lare; validation: Koffi Sagna, Amah Séna d'Almeida; formal analysis: Yendoubouame Lare, Koffi Sagna, Amah Séna d'Almeida; investigation: Yendoubouame Lare; resources: Koffi Sagna, Amah Séna d'Almeida; data curation, Yendoubouame Lare, Koffi Sagna, Amah Séna d'Almeida; writing original draft preparation, Yendoubouame Lare; writing review and editing: Koffi Sagna, Amah Séna d'Almeida; visualization, supervision: Koffi Sagna, Amah Séna d'Almeida; project administration: Koffi Sagna, Amah Séna d'Almeida; funding acquisition: Yendoubouame Lare, Koffi Sagna, Amah Séna d'Almeida. All authors reviewed the results and approved the final version of the manuscript.

Availability of Data and Materials: Not applicable.

Ethics Approval: Not applicable.

Conflicts of Interest: The authors declare no conflicts of interest to report regarding the present study.

Statement of Novelty: This work introduces a novel multi-order modeling framework to analyze the spatiotemporal evolution of velocity, density, and temperature fields within a carbon dioxide droplet exposed to supercritical conditions. Unlike previous studies that rely on simplified thermodynamic assumptions or macroscopic droplet behavior, this approach incorporates first- and second-order perturbation dynamics to resolve the internal structure of the transition

layer. The explicit separation of zeroth- and first-order contributions to heat flux and density evolution reveals non-trivial couplings between initial perturbations and the base state. These results offer new insights into droplet stability and phase transition mechanisms, with direct implications for modeling supercritical fluid behavior and optimizing propulsion systems. Controlling evaporation-induced discontinuities may significantly enhance fuel efficiency and combustion stability in high-performance applications.

Nomenclature

Symbol	Definition	Unit
r'	Radial distance from droplet center	m
$R'(t)$	Instantaneous droplet radius	m
$u'(r', t')$	Radial velocity component	$\text{m} \cdot \text{s}^{-1}$
t'	Time	s
T	Temperature	K
T_c	Critical temperature of CO ₂ (31.1°C)	K
P	Pressure	Pa
P_c	Critical pressure of CO ₂ (73.8 bar)	Pa
ρ	Density	$\text{kg} \cdot \text{m}^{-3}$
ρ_c	Critical density of CO ₂ (467 $\text{kg} \cdot \text{m}^{-3}$)	$\text{kg} \cdot \text{m}^{-3}$
C_p	Specific heat at constant pressure	$\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
C_v	Specific heat at constant volume	$\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
λ	Thermal conductivity	$\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$
R	Specific gas constant	$\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
a, b	Van der Waals constants	$\text{m}^6 \cdot \text{Pa} \cdot \text{mol}^{-2}$ and $\text{m}^3 \cdot \text{mol}^{-1}$
t_a	Acoustic time scale	s
t_{dif}	Diffusion time scale	s
M_a	Mach number	-
γ	Ratio of specific heats	-

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