Thermophysical analysis of microconfined turbulent flow regimes at supercritical fluid conditions in heat transfer applications

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ABSTRACT

The technological opportunities enabled by understanding and controlling microscale systems have not yet been capitalized to disruptively improve energy processes, especially heat transfer and power generation. The main limitation corresponds to the laminar flows typically encountered in microdevices, which result in small mixing and transfer rates. This is a central unsolved problem in the thermal-fluid sciences. Therefore, this work focuses on analyzing the potential of supercritical fluids to achieve turbulence in microconfined systems by studying their thermophysical properties. In particular, a real-gas thermodynamic model, combined with high-pressure transport coefficients, is utilized to characterize the Reynolds number achieved as a function of supercritical pressures and temperatures. The results indicate that fully-turbulent flows can be attained for a wide range of working fluids related to heat transfer applications, power cycles and energy conversion systems, and presenting increment ratios of $\mathcal{O}(100)$ with respect to atmospheric (subcritical) thermodynamic conditions. The underlying physical mechanism to achieve relatively high Reynolds numbers is based on operating within supercritical thermodynamic states (close to the critical point and pseudo-boiling region) in which density is relatively large while dynamic viscosity is similar to that of a gas. In addition, based on the Reynolds numbers achieved

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and the thermophysical properties of the fluids studied, an assessment of heat transfer at turbulent microfluidic conditions is presented to demonstrate the potential of supercritical fluids to enhance the performances of standard microfluidic systems by factors up to approximately $50\times$*.*

NOMENCLATURE

Latin letters

A_h Cross-sectional area [m^2]

c_p Isobaric specific heat capacity [J/(kg·K)]

h Heat convection coefficient [W/(m^2·K)]

K_a Association factor

L Length scale [m]

M Dipole moment [D]

Nu Nusselt number

P Pressure [Pa]

Pe Péclet number

P_h Cross-sectional perimeter [m]

Pr Prandtl number

Re Reynolds number

R_u Universal gas constant [J/(K·mol)]

T Temperature [K]

u Characteristic velocity [m/s]

$\bar{v}$ Molar volume [m^3/mol

W Molecular weight [kg/mol]

Z Compressibility factor

Greek symbols

Thermal diffusivity [m^2/s]

Thermal conductivity [W/(m·K)]

Dynamic viscosity [Pa·s]

Kinematic viscosity [m^2/s]

Density [kg/m^3]

$!$ Acentric factor

Subscripts

b Bulk value

c Critical point

1 INTRODUCTION

Microfluidics technology has grown rapidly over the past decades due to its high surface-to-volume ratios, flow and thermodynamic controllability, and length scales efficiently suited for interacting with microscopic elements [1]. These properties have proven to be well-suited to, especially, biology and chemistry, in which localized precision is usually an advantage. In general, microfluidic devices (i) can be easily miniaturized, enabling rapid analysis using portable instrumentation; and (ii) use minimal amounts of samples, reducing the waste generated and leading to overall low-cost operations. However, the utilization of microfluidics for energy applications, especially heat transfer and power generation (these consume, for example, an aggregated 70% of the European Union energy [2]), has long been a key challenge, in what some researchers refer to as "*lab-on-a-chip and energy — the microfluidic frontier*" [3]. One of the main reasons is that standard microdevices provide small rates of mixing and transfer, as a consequence of the laminar (smooth) flow regimes induced, resulting in significantly low efficiencies in comparison to macroscale systems in which turbulence is typically present. In particular, as a result of their small characteristic fluid velocities and sizes, the Reynolds numbers achieved in most standard microfluidic systems are in the order of $Re = uL = \sim 0{:}1 - 100$, with u and L the characteristic velocity and length scales and the kinematic viscosity of the fluid, and consequently flows are constrained to the laminar regime. The smooth nature of this flow regime does not provide good mixing efficiencies in comparison to the performances obtained if turbulence, which is characterized by rapid fluctuations of flow variables in time and space, is present [4]. As a result, standard microfluidic devices rely solely

on diffusive mixing, which is an inherently slow process. Nonetheless, as analyzed in this work, reaching supercritical fluid conditions in microdevices has the potential to enable a fine-controlled transition between laminar and turbulent regimes that could be leveraged to overcome the present limitations for a wide range of flow rates and microsystem sizes.

In most macroscale energy applications, for example related to power and heat transfer, turbulence is a key mechanism for achieving high levels of performance and efficiency due to the notable increase in mixing and transfer rates that it provides [5]. While there is no theorem relating *Re* to turbulence, flows at *Re* larger than $4000 - 5000$ are typically (but not necessarily) turbulent, while those at low *Re* usually remain laminar. In internal flows, for example, turbulence can first be sustained if *Re* is larger than a critical value of approximately 2300 [4]; as detailed in Section 2, turbulence is generally interspersed with laminar flow in confined geometries until a *Re* of about 4000 is achieved.

As introduced above, mixing in microdevices is one of the pivotal technologies for enabling miniaturized analysis and solutions. However, miniaturization also presents important challenges that do not occur in our everyday macroscopic world. In particular, the smooth and ordered flow structures caused by the dominant viscous effects makes achieving turbulence highly challenging. In fact, as it is commonly written in many textbooks and research publications: "*turbulent flow regimes used in macroscale applications to improve mixing is not possible at the microscale*" [1]; an important outcome of this work, therefore, is to change such *standardized* assumption through the utilization of supercritical fluids. As a result, most research in microfluidics is focused on improving molecular diffusion in laminar flows through different strategies [6], like for example fabricating serpentine-like microchannels. However, these techniques typically impose important construction complexities without exceptional increases in mixing and transfer rates. In general, typical microfluidic devices are characterized by hydraulic diameters in the range $D_h \sim 1-1000$ m and operate at average (bulk) velocities of $u_b \sim \mathcal{O}(1)$ m/s and smaller; significantly larger u_b have been reported in the literature, like for example 10 m/s [7] and 50 m/s [8], but these where exploration and/or testing scenarios under carefully controlled laboratory experiments and, thus, not envisioned for industrial/commercial applications. Consequently, standard fluids at atmospheric

conditions are not able to reach turbulent regimes in microfluidic systems as their kinematic viscosities are typically in the range $\sim 10^{-6} - 10^{-4}$ m^2/s, and consequently viscous forces dominate over inertial effects.

One of the most explored approaches for trying to achieve turbulent flow characteristics in microchannels is the use of electrokinetic forcing [9, 10]. For example, as argued by the authors, Wang et al. [11] have reported a direct observation of the existence of "*turbulence*" in microfluidics with $Re \sim 1$ in a pressure-driven flow under electrokinetic forcing using a novel velocimeter with ultrahigh spatiotemporal resolution. However, the chaotic flow and mixing induced corresponds to forced stirring. Low Reynolds numbers imply that viscous forces are more important than inertial destabilizing effects, and as a result the fluid is stirred but it does not develop turbulent mixing characteristics by itself; viz. there is no inherent multiscale spatio-temporal flow distribution. A different (brute-force) strategy is to extraordinarily increase the volumetric flow rate (large sizes/velocities) in microdevices to reach incipient turbulent flow conditions [8, 12, 13]. Although interesting for some applications, like for example high throughput liquid-liquid extraction and formation of emulsions [14], this approach presents two main drawbacks: (i) there is no intrinsic mechanism to trigger flow destabilization, and (ii) the volumetric flow rates required are notably high in contradiction to the small rates typically sought in microfluidic applications. Finally, an interesting approach recently proposed is the utilization of flexible microfluidics [15]. However, this technology has not yet been demonstrated at the microscale since the experiments were performed in microchannels of width in the order of millimeters.

In this work, instead, a drastically different approach based on directly developing turbulent flow in microfluidic systems by means of leveraging the properties of supercritical fluids is analyzed. High-pressure supercritical fluids (SCF) are substances at temperatures and pressures above their critical values (T_c, P_c). Fluids under such thermodynamic conditions do not separate into different phases. However, within the supercritical region, it is important to distinguish between supercritical gas-like and liquid-like fluids [16]: (i) a supercritical liquid-like fluid is one whose density is large, whose isothermal compressibility is small, and whose transport coefficients behave similar to a liquid; (ii) the density of supercritical gas-like fluids is smaller, their

isothermal compressibility is larger, and their transport coefficients vary similar to gases. At supercritical pressures, a smooth transition from liquid-like to gas-like supercritical behavior occurs at the pseudoboiling (also referred to as Widom) temperature T_{pb}. This temperature corresponds approximately to the point where a second-order (continuous) phase transition occurs, with the thermophysical properties rapidly changing. In particular, the strategy proposed makes use of this smooth transition when crossing the pseudo-boiling line to tune supercritical fluids to present liquid-like densities ($\sim 10^3$ kg/m^3) and gas-like viscosities ($\sim 10^{-5}$ Pa·s), and therefore achieve $Re \sim 10^3 - 10^4$ for typical microfluidic velocities and channel sizes favoring, in this manner, inertial over viscous forces and resulting in turbulent flow. As it will be investigated in this work, these properties could lead, without the need of additional mechanical micromixers or invasive strategies, to (i) a wide range of tunable Reynolds numbers spanning the laminar and turbulent regimes, (ii) faster mixing rates, (iii) enhanced mass, momentum, energy and species transfer, and (iv) smaller dispersion and residence time distributions. These properties have started to be studied [17] for flow-through chemistry, material science and deep-ocean biology; additional examples of microfluidic applications utilizing supercritical fluids (high pressure and temperature) include: (i) analyses of fluid properties of oil films and CO_2-oil diffusivity [18]; (ii) dew point studies of water-CO_2 mixtures [19]; and (iii) microwave-assisted catalytic combustion [20]. However, operating at high-pressure supercritical conditions to achieve turbulent regimes in microfluidic-based energy applications is a virtually uncharted research area. For example, Zhang *et al.* [21] have recently explored the idea of mixing intensification for antisolvent processes at microfluidic regimes of $D_h \approx 300$ and $u_b \approx 1.2 - 2.4$ m/s by means of utilizing high pressures in free-shear coflow configurations at isothermal conditions. Nonetheless, the overall strategy is significantly different to this work as: (i) jet flows are inherently unstable, and consequently laminar-to-turbulent transition occurs in the range $Re \approx 30 - 2000$ [22]; (ii) coflows require complex microfluidic configurations; and (iii) isothermal conditions are not suitable for energy-related applications.

This work, therefore, focuses on analyzing the potential of supercritical fluids to reach fully-developed turbulent regimes in microconfined flows with application to energy systems. The operating fluids considered are related to heat transfer, power cycles and energy conversion

systems [16, 23, 24, 25], and correspond to: CO_2, CH_4, N_2, O_2, R134a and H_2O. Additional commonly used operating fluids in energy applications correspond for example to: (i) H_2, C_6H_{14}, $C_{12}H_{26}$ and RP1 in high-pressure propulsion systems [16]; and (ii) R1233zd, R290 and R338ea in cooling/heating applications [25]. These supplemental fluids will not be studied in detail, but the insight obtained from this work can be straightforwardly generalized to a wide range of fluids utilized in energy systems, supercritical applications, and microfluidic devices. In this regard, the paper is organized as follows. First, in Section 2, a summary of bulk Reynolds numbers obtained from the literature characterizing flow regimes in internal flows is presented. Second, the thermophysical model utilized is described and validated against reference data in Section 3. Next, in Section 4, an analysis of the flow regimes obtained, characterization of thermophysical properties, and the implications in heat transfer applications is carefully performed and discussed. Finally, in Section 5, the work is concluded and future directions are proposed.

2 INTERNAL FLOW REGIMES

The motion of thermal-fluid flows can be described by a variety of dimensionless numbers [26]. Particularly to turbulent internal flows [4], the ratio between inertial and viscous forces is characterized by the bulk Reynolds number defined as

$$Re_b = \frac{D_h u_b}{}; \tag{1}$$

where $D_h = 4A_h$=P_h is the hydraulic diameter with A_h and P_h the cross-sectional area and perimeter of the flow, respectively, u_b is the bulk (average) velocity, and and are the density and dynamic viscosity of the fluid. At low Reynolds numbers, fluid motion tends to be dominated by laminar (sheet-like) structures, while at high-Reynolds-number flows tend to be turbulent (vortical-like). The Re_b transition interval in which flows separate between laminar and turbulent conditions is referred to as transitional regime, also termed critical Reynolds number region. In the laminar regime, the flow is ordered and organized in layers with no mixing between adjacent streamlines. Additionally, the flow recovers such state even in the presence of small disturbances as viscous

effects rapidly dissipate vortical and/or destabilizing structures. At transitional conditions, the flow is no longer uniform and eddies start to be generated which enhance flow mixing. However, the flow is interspersed with laminar regions and reorders itself downstream of a relatively small disturbance as inertial effects tend to be of the same importance as viscous dissipation. Finally, at fully-developed turbulent regimes, the flow mixing is actively enhanced due to the presence of energetic three-dimensional (3-D) eddies of different sizes.

Many turbulent flows are bounded (at least in part) by one or more solid surfaces, and consequently this work focuses on internal flows by considering three canonical geometries [4]: (i) fully-developed channel flow, (ii) fully-developed duct flow, and (iii) fully-developed pipe flow. The channel flow setup is a periodic passage constrained between two parallel flat plates sufficiently separated such that near-wall effects are negligible at the centerline and the statistics are independent of the spanwise direction, so that the flow is statistically symmetric about the mid-plane and predominantly in the streamwise direction with the average velocity varying only in the wall-normal direction. The duct flow problem is also periodic in the streamwise direction, but in this geometry the flow is confined to a rectangular-section tube. Even so, the turbulent statistics become similar to those of turbulent channel flow as Re_b increases to large values [27]. Finally, pipe flow is an akin case in which fluid motion is restricted to a tube of circular section, and with the time-averaged cross-section velocity profile symmetric with respect to the pipe axis. In particular, since the seminal experiment of Osborne Reynolds in 1883 [28], pipe flow has played an important role in the development and understanding of turbulent flow, especially for laminar-to-turbulent transition and skin-friction coefficient studies over smooth and rough walls [29, 30].

Focusing on the three canonical internal flows described above, Table 1 summarizes the Re_b values characterizing laminar, transitional and turbulent regimes obtained from theoretical, computational and/or experimental published studies and databases. The Re_b values listed in the table will be utilized in Section 4 to determine under which supercritical pressure and temperature ranges flows can achieve stable turbulent regimes in microconfined systems. In detail, the laminar regime is defined at values $Re_b \ . \ 1000 - 2000$ for all three cases, and with duct flow presenting the lower Re_b limit. Laminar-to-turbulent transition regimes correspond to ranges defined with

Table 1: Bulk Reynolds numbers Re_b indicating laminar, transitional and turbulent regimes for internal ows according to published studies

	Laminar	Transitional	Turbulent
Channel ow	$Re_b <$ 1350[4]	1350$< Re_b <$ 3000[4]	Re_b & 1800[4, 31]
Duct ow	$Re_b <$ 1077[32]	940$< Re_b <$ 1350[33] 1535$< Re_b$. 2742[7] 1900$< Re_b <$ 2200[12]	$Re_b >$ 1077[32] Re_b & 2742[7]
Pipe ow	$Re_b <$ 1800[8, 34] $Re_b <$ 2000[4, 31]	1500$< Re_b <$ 2500[1, 29] 1800. Re_b . 2500[8, 30] 2000$< Re_b$. 4000[4, 31]	$Re_b >$ 4000[4, 31] $Re_b >$ 5300[34, 35]

Table 2: Critical pressure P_c, temperature T_c, density $_c$, dynamic viscosity $_c$, and thermal diffusivity $_c$ of the substances studied [38]

	CO_2	CH_4	N_2	O_2	R134a	H_2O
P_c [MPa]	7:38	4:60	3:40	5:05	4:06	221
T_c [K]	304	191	126	155	374	647
$_c$ [kg/m^3]	467	163	313	436	512	322
$_c$ [Pa s]	3:40 10 ^{5}	1:51 10 ^{5}	1:88 10 ^{5}	3:38 10 ^{5}	3:74 10 ^{5}	5:62 10 ^{5}
$_c$ [mm^2/s]	8:67 10 ^{3}	1:63 10 ^{2}	1:23 10 ^{2}	9:10 10 ^{3}	9:49 10 ^{3}	1:47 10 ^{2}

signi cant disparity between literature sources as 1000 . Re_b . 4000, in which pipe ow is the setup with highest transition limit. Finally, turbulent regimes in all three geometries can be found at Re_b & 2000 5000.

3 THERMOPHYSICAL FRAMEWORK

Supercritical uids operate within high-pressure thermodynamic spaces in which intermolecular forces and nite packing volume effects become important; critical values of the substances studied are listed in Table 2. As a result, ideal-gas thermodynamic modeling assumptions [36] combined with Sutherland's law [37] for transport coef cients do not apply.

3.1 Real-gas thermodynamics

The thermodynamic space of solutions for the state variables pressure P, temperature T, and density of a single substance is described by an equation of state. One popular choice for systems at high pressures, which is used in this study, is the Peng-Robinson equation of state [39] written as

$$P = \frac{R_u T}{v \quad b} \quad \frac{a}{v^2 + 2bv \quad b^2}; \tag{2}$$

with R_u the universal gas constant, $v = W/$ the molar volume, and W the molecular weight provided in Table 3 for the substances studied. The coef cients a and b take into account real-gas effects related to attractive forces and nite packing volume, respectively, and depend on the critical temperatures T_c, critical pressures P_c, and acentric factors $!$ listed in Tables 2 and 3. They are de ned as

$$a = 0:457\frac{(R_u T_c)^2}{P_c}\left[1 + c\left(1 \quad \sqrt{T/T_c}\right)\right]^2 \quad \text{and} \quad b = 0:078\frac{R_u T_c}{P_c}; \tag{3}$$

where coef cient c is provided by

$$c = \begin{cases} 0:380 + 1:485! \quad 0:164!^2 + 0:017!^3 & \text{if } ! > 0:49; \\ 0:375 + 1:542! \quad 0:270!^2 & \text{otherwise:} \end{cases} \tag{4}$$

The Peng-Robinson real-gas equation of state needs to be supplemented with the corresponding high-pressure thermodynamic variables based on departure functions [40] calculated as a difference between two states. In particular, their usefulness is to transform thermodynamic variables from ideal-gas conditions (low pressure - only temperature dependant) to supercritical conditions (high pressure). The ideal-gas parts are calculated by means of the NASA 7-coef cient poly-

Table 3: Molecular weight W, acentric factor ! , association factor $_a$, and dipole moment M of the substances studied [37]

	CO_2	CH_4	N_2	O_2	R134a	H_2O
W [kg/mol]	4:40 10 ^{2}	1:60 10 ^{2}	2:80 10 ^{2}	3:20 10 ^{2}	1:02 10 ^{1}	1:80 10 ^{2}
! [-]	2:24 10 ^{1}	1:14 10 ^{2}	3:72 10 ^{2}	2:22 10 ^{2}	3:27 10 ^{1}	3:44 10 ^{1}
$_a$ [-]	0.0	0.0	0.0	0.0	0.0	7:60 10 ^{2}
M [D]	0.0	0.0	0.0	0.0	2.06	1.85

nomial [41], while the analytical departure expressions to high pressures are derived from the Peng-Robinson equation of state as detailed in Jofre & Urzay [16]. Among the different variables, the speci c heat capacity at constant pressure c_p is the physical quantity utilized in the analyses conducted in this work.

The real-gas framework described above is validated in terms of density normalized by critical density $= {}_c$ against reference data from NIST [38] in Figure 1 at pressures $P=P_c = 0:1; 1; 10$ and temperatures $0:5 < T=T_c < 4:5$ for the substances studied. In general, except for low temperatures at $P=P_c = 0:1$ since the high-pressure model selected is derived and intended to be utilized at supercritical states, the plots show signi cant good agreement between reference data and model results for the pressure and temperature range considered. Nonetheless, except for R134a, minor discrepancies can be observed (especially for H_2O at $P=P_c = 10$) at low temperatures (liquid-like phase); similar discrepancies have been found in other works [42, 43, 16]. In this regard, volume-translating corrections to the Peng-Robinson equation (Eq. 2) exist that provide a higher accuracy of the liquid-like density near the critical point [44], but are not considered in the present study since they lead to a mismatch of the thermophysical variables (e.g., speci c heat), which is an undesirable characteristic for describing the transfer of heat across the pseudo-boiling region. However, these differences are relatively small [42, 43] and within the same order of magnitude, and consequently do not present important impacts on the thermophysical analyses that will be performed in Section 4.

(a) (b)

(c) (d)

(e) (f)

Fig. 1: Comparison between NIST data [38] (symbols) and thermophysical model (lines) of density normalized by the critical point value $= {}_c$ at pressures $P = P_c = 0:1; 1; 10$ and temperature range $0:5 < T = T_c < 4:5$ for CO_2 (a), CH_4 (b), N_2 (c), O_2 (d), R134a (e), and H_2O (f)

3.2 High-pressure transport coef cients

The high pressures involved in the analyses conducted in this work prevent the use of simple relations for the calculation of the dynamic viscosity and thermal conductivity . In this regard, standard methods for computing these coef cients for Newtonian uids are based on the correlation expressions proposed by Chung et al. [45, 46]. These correlation expressions are mainly function of critical temperature T_c and density $_c$ provided in Table 2, molecular weight W, acentric factor !, association factor $_a$ and dipole moment M listed in Table 3, and the NASA 7-coef cient polynomial [41]; further details can be found in dedicated works, like for example [16, 37].

Similar to the previous subsection, the high-pressure transport coef cients framework described above is validated in terms of dynamic viscosity normalized by the critical value $= _c$ against reference data from NIST [38] in Figure 2 at pressures $P=P_c = 0:1; 1; 10$ and temperatures $0:5 < T=T_c < 4:5$ for the substances studied. Similar to the previous subsection, except for low temperatures at $P=P_c = 0:1$ where the high-pressure model selected is not intended to be utilized, the plots show notable agreement between reference data and model results for the pressure and temperature range considered. Nonetheless, except for CO_2, small differences can be noticed (especially for H_2O at $P=P_c = 10$) at low temperatures (liquid-like phase). Nevertheless, these differences are relatively small and within the same order of magnitude, and consequently do not incorporate important discrepancies in the thermophysical analyses that will be conducted in Section 4. Hence, the high-pressure transport coef cients framework proposed is accurate enough in the thermodynamic space of supercritical temperatures and pressures studied [16, 43].

4 RESULTS & DISCUSSION

This section analyzes the potential of supercritical uids to achieve turbulent ow regimes in micro uidic-based energy applications by means of utilizing the thermophysical framework described in Section 3.

4.1 Flow regimes at supercritical conditions

Flow regimes at supercritical conditions are analyzed for CO_2, CH_4, N_2, O_2, R134a and H_2O. The supercritical ranges studied correspond to $1 < P=P_c < 100$ and $0:5 < T=T_c < 4:5$, and differ-

(a) (b)

(c) (d)

(e) (f)

Fig. 2: Comparison between NIST data [38] (symbols) and thermophysical model (lines) of dynamic viscosity normalized by the critical point value $= {}_c$ at pressures $P=P_c = 0:1; 1; 10$ and temperature range $0:5 < T=T_c < 4:5$ for CO_2 (a), CH_4 (b), N_2 (c), O_2 (d), R134a (e), and H_2O (f)

ent values of hydraulic diameter $D_h = 100; 200; 500$ m and bulk velocity $u_b = 0.5; 1.0; 2.0; 5.0$ m/s are considered. The maximum bulk Reynolds number (de ned in Eq. 1) obtained for each micro uidic setup is provided in Table 4. For all substances, these values are found (approximately) in the vicinity of their critical points. Five main observations can be inferred from the values listed in the table. First, independently of the geometrical setup and substance considered, micro uidic devices at supercritical conditions with $D_h = 100$ m and $u_b = 0.5$ m/s are indubitably restricted to laminar regimes as the maximum bulk Reynolds numbers attained are smaller than the limiting values presented in Table 2. However, transitional and turbulent regimes can be potentially achieved with the same D_h by increasing u_b by factors of 2 and 10 , respectively. Second, the scenario rapidly changes when increasing the hydraulic diameter to $D_h = 200$ m as the Re_b values obtained correspond to transitional ($u_b = 0.5; 1.0$ m/s) and turbulent regimes ($u_b = 2.0; 5.0$ m/s). Third, with $D_h = 500$ m, the ow can be considered to be turbulent for the three bulk velocities ($u_b = 0.5; 1.0; 5.0$ m/s) and substances considered. Fourth, it is important to notice that turbulent ows at relatively high Reynolds numbers of roughly Re_b 15000 and Re_b 40000 can be achieved in micro uidic systems at supercritical uid conditions with $D_h = 200; 500$ m, respectively, and $u_b = 5.0$ m/s. Fifth, the setup with $D_h = 200$ m and $u_b = 2.0$ m/s has been highlighted as it represents a popular con guration in a wide range of micro uidic applications in which fully-developed turbulent regimes could be potentially achieved by utilizing supercritical uids.

The row highlighted in Table 4 indicates a particular set of micro uidic conditions ($D_h = 200$ m and $u_b = 2.0$ m/s) that will be studied in detail in this work as a reference setup. In particular, Figure 3 depicts color maps with iso-contour lines of the estimated bulk Reynolds numbers for the substances considered at this micro uidic reference con guration within the supercritical region de ned by $1 < P/P_c < 100$ and $0.5 < T/T_c < 4.5$; critical points and pseudo-boiling curves are indicated by red diamonds and violet dashed-dotted lines. Focusing on the regions in which turbulent ow can be achieved for the internal ows considered, three main color distributions can be identi ed: (i) CO_2, CH_4, N_2 and O_2 present concave-type teardrop shapes that increase width along the pseudo-boiling line (especially above) starting from the critical point; (ii) R134a shows a triangular-like shape that grows toward lower temperatures as pressure increases; and (iii) H_2O

Table 4: Maximum bulk Reynolds numbers Re_b and for the substances studied at different hydraulic diameters D_h and bulk velocities u_b. These maximum Reynolds numbers are found (approximately) at the critical point for each substance

D_h [m]	u_b [m/s]	CO_2	CH_4	N_2	O_2	R134a	H_2O
100	0.5	659	486	816	841	796	664
100	1	1317	973	1631	1682	1592	1328
100	5	6587	4864	8156	8412	7961	6641
200	0.5	1317	973	1631	1682	1592	1328
200	1	2635	1945	3263	3365	3185	2657
200	2	5269	3891	6525	6730	6369	5313
200	5	13174	9727	16313	16825	15923	13283
500	0.5	3293	2432	4078	4206	3981	3321
500	1	6587	4864	8156	8412	7961	6641
500	5	32934	24318	40781	42062	39807	33206

displays an almost horizontal region of larger Re_b values below the critical temperature. In general, based on the limiting values provided in Table 1, turbulent ow regimes are achieved in most of the supercritical region studied for the six substances, except for relatively (i) large temperatures - low pressures and (ii) large pressures - low temperatures. In fact, the supercritical regions achieving turbulent ow characteristics are signi cantly robust as they present relatively wide P-T intervals.

The corresponding density and dynamic viscosity distributions normalized by their critical values are shown in Figures 4 and 5, respectively. The density distributions are similar for the substances studied: (i) they present quarter-circle arc shapes increasing from 0.1 to 4 in the direction toward higher pressures and lower temperatures, and (ii) the larger variations are strongly concentrated around the pseudo-boiling region. The distributions of normalized dynamic viscosity for CO_2, CH_4, N_2 and O_2 depict similar shape topologies as for density, but ranging wider ratios of order 10^{-1} 10^{3} with rapid variations shifted toward lower temperatures and slightly larger pressures. Substances R134a and H_2O display signi cantly different distributions characterized by: (i) smaller ratios of dynamic viscosity, especially H_2O; and (ii) distinct topologies with ratios below 1 notably concentrated around the pseudo-boiling region, with H_2O presenting larger values

(a) (b)

(c) (d)

(e) (f)

Fig. 3: Bulk Reynolds numbers Re_b in the range $1 < P=P_c < 100$ and $0.5 < T=T_c < 4.5$ with $D_h = 200$ m and $u_b = 2$ m/s for CO_2 (a), CH_4 (b), N_2 (c), O_2 (d), R134a (e), and H_2O (f). Critical points and pseudo-boiling curves are indicated by red diamonds and violet dashed-dotted lines, respectively

at high supercritical temperatures and pressures in comparison to the other substances, providing, thus, an explanation for the signi cantly larger bulk Reynolds numbers this uid obtains at low $T=T_c$ values. In consequence, it is important to highlight that, as shown by these two sets of color map distributions, liquid-like to gas-like transitions across the pseudo-boiling region for density and dynamic viscosity do not occur exactly at the same P-T values, resulting in the opportunity to tune supercritical uids to present liquid-like densities combined with gas-like viscosities to achieve turbulent ow regimes in micro uidic systems. In particular, transitioning from liquid-like to gas-like supercritical thermodynamic conditions, density reductions are concentrated in a narrow region around the pseudo-boiling line, while increments in dynamic viscosity are more spread and shifted toward lower temperatures. Additionally, the liquid-like to gas-like transition observed is smooth for the substances studied, indicating that the tunability of supercritical uids can be achieved with signi cant controllability and precision in practical applications.

4.2 Characterization of thermophysical properties

The connection between ow regimes and thermophysical properties at supercritical conditions is studied rst by analyzing the compressibility factor $Z = PW/(R_u T)$ color maps shown in Figure S1 (available in Supplemental Material); viz. $Z \neq 1$ indicate deviations from ideal-gas thermodynamic conditions. For the six substances studied, the largest deviations for $Z > 1$ (densities smaller than the ideal-gas corresponding values) are found at supercritical high pressures and low temperatures with values $Z \quad 10$, whereas departures toward $Z < 1$ (densities larger than the ideal-gas corresponding values) are located at supercritical temperatures below the critical point for $P=P_c$. 5; notice that the color maps look topologically similar as Z is an inherent property of each equation of state model. Nonetheless, it is interesting to highlight that the supercritical thermodynamic ranges in which turbulent ow regimes can be achieved at micro uidic conditions, see Figure 3, involve compressibility factors in the range 0:1 . Z . 5, and consequently real-gas effects in density are not the only factors dictating the ow regime, it is instead the combination of despair variations of density and dynamic viscosity across the pseudo-boiling region.

The relationship between heat transfer and thermophysical properties within the supercritical thermodynamic region de ned by $1 < P=P_c < 100$ and $0:5 < T=T_c < 4:5$ is studied next in terms

(a) (b)

(c) (d)

(e) (f)

Fig. 4: Ratios of density to critical density $= {}_c$ in the range $1 < P = P_c < 100$ and $0.5 < T = T_c < 4.5$ for CO_2 (a), CH_4 (b), N_2 (c), O_2 (d), R134a (e), and H_2O (f). Critical points and pseudo-boiling curves are indicated by red diamonds and violet dashed-dotted lines, respectively