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On the importance of non-equilibrium models for describing the coupling of heat and mass transfer at high pressure

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ABSTRACT

The present work summarises the main findings of a systematic study on the behaviour of liquid droplets embedded in a high pressure and temperature environment. Thanks to the availability of quantitative temperature data, it was possible for the first time to assess the conceptual soundness of different theoretical approaches. In all cases, the predictions from non-equilibrium models agree fairly well with Raman-extracted temperatures. By performing a thermodynamic analysis of interfacial transport in the framework of the Onsager theory, the following was found. Models that assume vapour-liquid equilibrium fail to provide a good estimation of the droplet temperature. This is due to the description of the interfacial fluxes that does not include the interdependency between heat and mass transfer, as prescribed by the Onsager theorem. Under non-equilibrium conditions, interfacial temperature jumps may exist without leading to the disintegration of the immiscible interface, as corroborated by several theoretical and experimental studies. The inception of thermal losses is controlled by the interfacial resistivities rather than by the Knudsen number. With decreasing droplet size and increasing ambient temperatures, the inclusion of local non-equilibrium effects in the modelling of the interfacial fluxes from the vapour side leads to higher interfacial temperature jumps.

1. Introduction

The study of near-critical fluid injection has received increasing attention in the last decade, as testified by the large body of literature recently published on the subject [1–6]. Unfortunately, the understanding of the mixing process in high-pressure, heterogeneous media is still very controversial, due to the strong coupling between the transport of energy, mass and momentum across interfaces. Crua et al. [7] recently proposed a classification of mixing processes for droplets at Diesel engine conditions, based on the analysis of shadowgrams. The authors identified three regimes, namely classical evaporation, transitional and diffusive mixing. A striking feature is that the criterion proposed for the onset of the different mixing regimes is solely a function of the ambient gas pressure and temperature, normalised with respect to the critical values of the injected fluid. This seems to suggest that fuel properties and initial conditions are sufficient to characterise the mixing behaviour at high pressure conditions. It is arguable, however, whether this classification is not biased by the three dimensional trajectory of droplets emitted from Diesel injectors. As the droplet moves away from the focal plane of the optical setup, the images become increasingly blurred and only the wake becomes visible. This is particularly critical at high ambient pressures and temperatures, where it becomes increasingly difficult to discern micro-sized droplets moving away from the focal plane due to their reduced initial size. A different interpretation was proposed by Newman and Brzustowski [8]. The authors pointed out that the presence of gaseous regions within a shadowgram cannot be automatically associated to the onset of diffusive mixing, since one cannot exclude evaporation as the leading mechanism for mixture formation. Consequently, they repeated the experiments in a fuel-saturated atmosphere, in order to quench evaporation and enable a transition to dense fluid mixing. Their findings were later corroborated by the experiments of Chehroudi et al. [9], who observed a transition to a supercritical jet when liquid nitrogen was injected into a (partially) saturated atmosphere. The important insight from Newman and Brzustowski's work is that the evolution of jets (or droplets) in a high pressure environment is controlled by the local energy and mass fluxes rather than by the initial ambient conditions.

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Nomenclature

Acronym

EoS	Equation of State
EQ	Equilibrium
NEQ	Non-Equilibrium
PC-SAFT	Perturbed-Chain Statistical Associating Fluid Theory
VLE	Vapour Liquid Equilibrium

Greek Symbol

α_T	binary thermal diffusion factor, [mol/kg]
γ	surface tension, [N/m]
κ	thermal conductivity, [W/(m K)]
μ	chemical potential, [J/mol]
ρ	density, [kg/m^3]
τ	viscous stress, [N/m^2]
ζ	local interfacial resistivity, [m/(W K)] or [m s/(W K)]

Roman

$\dot{E}$	energy flow rate, [J/s]
$\dot{m}$	mass flow rate, [kg/s]
Kn	Knudsen number, [−]
c	concentration, [mol/m^3]
c_p	specific heat capacity at constant pressure, [J/(kg K)]
e	specific internal energy, [J/kg]
h	specific enthalpy, [J/kg]
J	diffusive flux, [kg/(m^2 s)] or [J/(m^2 s)]
L	latent heat, [J/kg]
l_{VLE}	interface thickness, [m]
L_{ij}	phenomenological coefficients
l_{th}	length scale for thermal diffusion, [m]
m	mass, [kg]
M_i	species molar mass, [kg/mol]
p	pressure, [Pa]
R	total interfacial resistivity, [m^2/(W K)] or [m^2 s/(W K)]
r	radius, [m]
R_u	universal gas constant, [J/(K mol)]
T	temperature, [K]
t	time, [s]
u	velocity, [m/s]
v	specific volume, [m^3/kg]
x	mole fraction, [−]
z	coordinate along the surface thickness, [m]

Subscripts

sat	saturated
ch	chamber
d	droplet
e	specific internal energy
f	fluid
g	gas
i	Knudsen-continuum interface
l	liquid
m	mass
mix	mixture
q	heat
s	surface
v	vapour

At theoretical level, two distinct conceptual approaches have been proposed, namely vapour-liquid equilibrium (VLE) and non-equilibrium (NEQ) formulations. The latter encompass a detailed modelling of the fluxes across interfaces in heterogeneous media, in compliance with the theorems of non-equilibrium thermodynamics. As such, NEQ models allow the thermodynamic system "droplet" (or jet) to exist also under unstable thermodynamic conditions, albeit only temporarily. Relaxation towards equilibrium occurs then through simultaneous mass (J_m) and energy (J_e) fluxes across its interface. Examples of NEQ approaches can be found in the work of Bellan's group [10,11] on the evolution of supercritical mixing layers and in Young [12] or Kjelstrup and Bedeaux [13] on the evaporation/condensation of spherical droplets. VLE formulations, instead, restrict the evolution of the thermodynamic system "droplet" to a set of stable thermodynamic states, compatible with the chosen equation of state (EoS). In recent years, the use of equilibrium frameworks to describe the evolution of high-pressure fluids in presence of large temperature and vapour pressure (or concentration) gradients has become increasingly common (see e.g. [14–16]), while the further development of non-equilibrium models has received much less attention.

The fundamental question addressed in this work is to investigate whether modelling approaches that assume vapour-liquid equilibrium are suited to predict the evolution of fluid dynamic processes occurring at large departure from equilibrium. Since modern engines operate at increasingly higher pressure and temperature conditions, this question is both legitimate and central to the current scientific discourse. According to Sonninno [17], for systems subject to large gradients of thermodynamic quantities (e.g. temperature), it is necessary to extend the transport closure equations by including nonlinear corrections to the linear "Onsager" transport coefficients. Unfortunately, due to the paucity of quantitative data, it was not possible until now to evaluate the different theoretical models and the assumptions that underpin them. As a first contribution to the ongoing debate, we perform a systematic study on the evolution of single droplets, embedded in a hot, pressurised nitrogen atmosphere. Raman measurements, performed in the droplet wake, are processed to provide an estimate of the droplet surface temperature. These values are then compared to the predictions from different theoretical models. It is found that VLE models systematically underestimate the droplet temperature over a large part of its lifetime. In order to explain this discrepancy, a thermodynamic analysis of the interfacial transport processes is performed.

2. Methodology

2.1. Raman measurements

The experiments are performed in a stainless steel chamber that can be operated up to 60 bar and 553 K. The temperature is monitored by type-T thermocouples at different locations with an accuracy of ± 1 K. PID-controllers regulate six resistance heater rods, located in the vertical walls and in the horizontal base, in order to assure a homogeneous temperature distribution. The droplet generator is mounted in the top lid, whose temperature is controlled by a thermo-oil bath up to a maximum value of 508 K. The temperature is measured with a resistance thermometer Pt100 with a class A accuracy [$\pm (0.15 + 0.002|T|)^{\circ}$C]. The light source is a frequency doubled Nd:YAG laser, emitting two consecutive pulses of 0.3 J with a 2 μs delay between them and an individual pulse length of 7.5 ns. Fig. 1(a) shows schematically the Raman optical setup. A polarising beam splitter regulates the intensity of the laser beam that is transmitted to the light sheet optics. The Region-Of-Interest (ROI) is also schematically illustrated in Fig. 1(b) (dashed box). The measurement volume is imaged using two