



**HAL**  
open science

# A multicomponent real-fluid fully compressible four-equation model for two-phase flow with phase change

Ping Yi, Songzhi Yang, Chaouki Habchi, Rafael Lugo

► **To cite this version:**

Ping Yi, Songzhi Yang, Chaouki Habchi, Rafael Lugo. A multicomponent real-fluid fully compressible four-equation model for two-phase flow with phase change. *Physics of Fluids*, 2019, 31 (2), pp.026102. 10.1063/1.5065781 . hal-02061996

**HAL Id: hal-02061996**

**<https://ifp.hal.science/hal-02061996>**

Submitted on 8 Mar 2019

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1 A multicomponent real-fluid fully compressible four-equation model for two-phase flow with phase  
2 change

3  
4 Ping Yi<sup>1,2</sup>, Songzhi Yang<sup>1,2</sup>, Chaouki Habchi<sup>\*1,2</sup>, Rafael Lugo<sup>1</sup>

5 <sup>1</sup> IFP Energies nouvelles, 1 et 4 avenue de Bois-Préau, 92852 Rueil-Malmaison, France

6 <sup>2</sup> Institut Carnot IFPEN Transports Energies, 1 et 4 avenue de Bois-Préau, 92852 Rueil-Malmaison,  
7 France

8  
9 Abstract

10 A fully compressible four-equation model for multicomponent two-phase flow coupled with a real-  
11 fluid phase equilibrium-solver is suggested. It is composed of two mass, one momentum, and one  
12 energy balance equations under the mechanical and thermal equilibrium assumptions. The  
13 multicomponent characteristics in both liquid and gas phases are considered. The thermodynamic  
14 properties are computed using a composite equation of state (EoS), in which each phase follows its  
15 own Peng-Robinson (PR) EoS in its range of convexity, and the two-phase mixtures are connected  
16 with a set of algebraic equilibrium constraints. The drawback of complex speed of sound region for  
17 the two-phase mixture is avoided using this composite EoS. The phase change is computed using a  
18 phase equilibrium-solver, in which the phase stability is examined by the Tangent Plane Distance  
19 (TPD) approach; an isoenergetic-isochoric (UVn) flash including an isothermal-isobaric (TPn) flash is  
20 applied to determine the phase change. This four-equation model has been implemented into an in-  
21 house IFP-C3D software. Extensive comparisons between the four-equation model predictions,  
22 experimental measurements in flash boiling cases, as well as available numerical results were carried  
23 out, and good agreements have been obtained. The results demonstrated that this four-equation model  
24 can simulate the phase change and capture most real-fluid behaviors for multicomponent two-phase  
25 flows. Finally, this validated model was applied to investigate the behaviors of *n*-dodecane/nitrogen  
26 mixtures in one-dimensional shock and double-expansion tubes. The complex wave patterns were  
27 unraveled, and the effects of dissolved nitrogen and the volume translation in PR EoS on the wave  
28 evolutions were revealed. A three-dimensional transcritical fuel injection is finally simulated to  
29 highlight the performance of the proposed four-equation model for multidimensional flows.

30  
31 Keywords: Multicomponent two-phase flow, Four-equation model, Real fluid, Phase change, Peng-  
32 Robinson EoS, Flash boiling

33  
34 1. Introduction

35 Accurate and robust modeling of compressible two-phase flow is crucial for many engineering  
36 applications, such as fuel injectors, nuclear reactors, rocket motors, as well as gas turbines and heat

1 pumps (Menter, 1994). The involved two-phase flow maybe subcritical, transcritical or supercritical  
2 depending on the pressure and temperature operating conditions. Some subsonic, sonic, or supersonic  
3 regions may appear due to shock and expansion waves (Courant and Friedrichs, 1999). Indeed, these  
4 phenomena are characterized by the violent variations of local Mach number, the large density  
5 gradient across liquid and gas phases, the intense compressibility and gas solubility effects, as well as  
6 the phase change and non-ideal thermodynamic properties (Yang et al., 2017). Especially, the phase  
7 change adds more complexity to the two-phase flow simulation, such as cavitation, strong shock or  
8 flash boiling phenomena. Several numerical models have been developed for the simulation of such  
9 two-phase flow, with the numbers of transport equations ranging from three to seven depending on the  
10 initial equilibrium assumptions. One of the difficulties in modelling is the physical transfer process  
11 taking place across the phases interfaces, such as mass, momentum and heat transfer.

12  
13 The most general two-phase flow model is the fully non-equilibrium seven-equation model, in which  
14 each phase has its own pressure, velocity and temperature, and is governed by its own set of fluid  
15 equations. More precisely, it is based on a fully compressible model composed of three balance  
16 equations for the gas phase and three balance equations for the liquid phase, together with a transport  
17 equation for the phase volume fraction. Such a non-equilibrium model is built using relaxation  
18 methods with finite characteristic time for velocity, pressure, temperature and chemical potential at the  
19 phase interface (Baer and Nunziato, 1986, Flåtten and Lund, 2011). Alternatively, the stiff relaxation  
20 approaches have been proven to be numerically stable. For instance, the pressure and velocity between  
21 two phases can be relaxed instantaneously (Saurel and Abgrall, 1999), and these stiff relaxation  
22 procedures have also been applied to the temperature and Gibbs free energy (Habchi, 2015, Zein et al.,  
23 2010). The seven-equation model shows great capabilities in describing complex wave patterns and  
24 correctly capturing the wave propagation in liquid and gas phases, separately (Andrianov et al., 2003).  
25 However, the complexity of implementation in computational fluid dynamics software has limited its  
26 extensive use, and simpler models are often preferred.

27  
28 In order to simplify the seven-equation model, the reduced five-equation models, in which the  
29 mechanical and thermal equilibrium are assumed, have been proposed extensively (Allaire et al., 2002,  
30 Kapila et al., 2001, Murrone and Guillard, 2005, Saurel et al., 2009). Kapila et al. (2001) have  
31 constructed the most popular formulation with two mass conservation equations for the liquid and  
32 vapor, one mixture momentum equation, one mixture energy equation, together with a transport  
33 equation for the liquid volume fraction. It has been demonstrated that this model shows excellent  
34 resolution of interfaces between two compressible fluids (Murrone and Guillard, 2005, Petitpas et al.,  
35 2007). However, serious numerical oscillations have been observed due to the non-conservative  
36 feature of liquid volume transport equation in this five-equation model. Some other simplified two-

1 phase flow models derived from the above general seven-equation model by assuming zero relaxation  
 2 time have been proposed and developed (Ransom and Hicks, 1984, Troshko and Hassan, 2001), and  
 3 the details of these models are summarized in the work of (Zein et al., 2010).

4

<b>Nomenclature</b>			
$a, b$	Coefficients in PR EoS	$\psi$	Phase molar fraction
$A, B$	Coefficients in PR EoS	$\phi$	Fugacity coefficient
$C_{s,p}$	Speed of sound for each phase	$\varepsilon$	Tolerance
$C_{s,mix}$	Speed of sound for two-phase	$\Delta t$	Time-step
$C_p$	Isobaric heat capacity	<i>Superscript</i>	
$e$	Internal energy	*	Specified constant values in UVn flash
$f_k$	Fugacity	"	Non-equilibrium values from flow-solver
$f_a$	Positive real number	$L$	Laminar
$F_{1,2,3,4}$	Objective functions	$T$	Turbulent
$J$	Flux in control volume	<i>Subscript</i>	
$k$	Binary interaction coefficient	$p$	Phase index
$K$	Equilibrium factor	$k$	Species index
$\dot{m}$	Phase change mass rate	$mix$	Mixture
$M$	Molecular weight	$N$	Total number of species
$n_k$	Mole number of species	$g$	Gas
$R$	Universal gas number	$l$	Liquid
$T$	Temperature	$L$	Left
$t$	Time	$R$	Right
$P$	Pressure	$0$	Initial values
$Pr$	Prandtl number	<i>Abbreviation</i>	
$q$	Conduction heat flux	$CFL$	Courant-Friedrichs-Lewy
$u$	Velocity	$C_{12}$	N-dodecane
$U_F$	Front velocity of evaporation wave	$EoS$	Equation of state
$U_L, U_R$	Initial conditions	$H_2O$	Water
$V$	Molar volume	$MM$	Multicomponent in both liquid and gas
$x, y$	Compositions in liquid and gas phase	$NASG$	Noble-Abel-Stiffened Gas
$Y$	Mass fraction	$N_2$	Nitrogen
$z$	Feed ( $z_k = n_k / \sum n_k$ )	$PR$	Peng-Robinson
$Z$	Compressibility factor	$QSOU$	Quasi Second Order Upwind
<i>Greek letters</i>		$SG$	Stiffened Gas
$\alpha$	Phase volume fraction	$SM$	Single component in liquid phase, and multicomponent in gas phase
$\rho$	Density	$TPD$	Tangent Plane Distance
$\tau$	Stress tensor	$TPn$	Isothermal-isobaric flash
$\lambda$	Heat conductivity	$UVn$	Isoenergetic-Isochoric flash
$\mu$	Dynamic viscosity	$1D, 3D$	One-, three-dimension
$\omega$	Acentric factor		

5

1 Three and four-equation models are also very prevalent and have been widely used to simulate the  
2 cavitating flows (Habchi et al., 2008, Kunz et al., 2000, Moreau et al., 2004, Venkateswaran et al.,  
3 2002). Four-equation models are composed of three conservation laws for mixture quantities (mass,  
4 momentum, energy) in addition to eventual partial density transport for multicomponent problems,  
5 along with a phase change source term in the right-hand side. Four-equation models have been proven  
6 to show high numerical efficiency (Battistoni et al., 2014). However, the main difficulties of this kind  
7 of model are to estimate the mass source term of phase change and the tunable parameters for the  
8 evaporation and condensation processes (Utturkar et al., 2005). Recently, (Saurel et al., 2016)  
9 proposed a new four-equation model, in which each phase is compressible and the two phases share  
10 common pressure, velocity, temperature, and Gibbs free energy. Moreover, a specific phase  
11 equilibrium-solver by applying the Noble-Abel-Stiffened Gas equation of state (Le Métayer and  
12 Saurel, 2016). Indeed, this fitted-parameters NASG EoS is used to simplify the thermodynamic  
13 computations. Distinct advantages of this four-equation model in conjunction with the phase  
14 equilibrium-solver have been shown in computational efficiency and numerical stability. It is worth  
15 noting that, this phase equilibrium-solver with NASG EoS is currently limited to the liquid phase only  
16 with single-component (i.e., gas solubility in liquid phase is neglected). In reality, however, substantial  
17 amount of gas dissolved in liquid phase under high pressure conditions makes primary influence on  
18 the nucleation and phase change in several industrial devices. Therefore, the full multicomponent real-  
19 fluids characteristics need to be considered.

20

21 The cubic equation of state (EoS) is well known to be able to capture the real-fluid behaviors for two-  
22 phase flow. The non-linearity of real-fluid may cause spurious pressure oscillations if it is not properly  
23 resolved (Banuti, 2015). Besides, the squared sound speed predicted by the cubic EoS (e.g., van der  
24 Waals) may become negative inside the spinodal region (see Fig. 1), which results in a loss of  
25 hyperbolicity (Menikoff and Plohr, 1989, Petitpas et al., 2009). Both Peng-Robinson (PR) (Peng and  
26 Robinson, 1976) and van der Waals EoSs belong to cubic EoSs holding the same repulsive term, and  
27 the PR EoS only improves the attractive term. Therefore, they hold similar fundamental properties.  
28 Even though (Menikoff and Plohr, 1989) only demonstrated the loss of hyperbolicity for van der  
29 Waals EoS for instance, the PR EoS holds the same drawback, which has been demonstrated by many  
30 scientific materials (Ma et al., 2017, Petitpas et al., 2009, Saurel et al., 2016, Saurel et al., 2017). All  
31 these fundamental drawbacks have limited the extensive applications of cubic EoSs.

32

33 Recently, the PR EoS has been used to simulate fuel injection processes, but mostly for the simulation  
34 of dense-gas or dense-liquid without phase change (i.e., transcritical conditions). For instance, the PR  
35 EoS and Soave-Redlich-Kwong EoS (Soave, 1972) have been used for the cryogenic flows (Schmitt et  
36 al., 2009, Terashima et al., 2013). The ECN spray A has also been simulated with the assumption of

1 transcritical conditions and no phase change (Ma et al., 2017), as well as considering the phase change  
2 (Matheis and Hickel, 2018). Most studies, however, only focused on the two- and three-dimensional  
3 (2D or 3D) simulations, the complex wave patterns in one-dimensional (1D) tube, up to now, have not  
4 been understood clearly with the real-fluid equilibrium-solver. The recent studies in one-dimensional  
5 tubes (Chiapolino et al., 2017, Goncalvès and Charrière, 2014, Zein et al., 2010) are mainly focusing  
6 on the simulation of water-nitrogen with fitted parameter EoSs, and only consider a single-component  
7 in the liquid phase (the dissolved gas is neglected). These studies cannot capture the real-fluid  
8 behaviors for multicomponent two-phase flows. Therefore, as a prelude of further 3D applications like  
9 diesel and gasoline injection modeling, thorough validations of multicomponent real-fluid two-phase  
10 flow model are necessary. This is carried out in the present study based on the 1D shock tube, flash  
11 boiling and double-expansion tube cases using *n*-dodecane/nitrogen mixture which are typical  
12 surrogates for fuel injection in internal combustion engines.

13

14 For the simulation of two-phase subcritical flows, a phase equilibrium-solver is needed, in which the  
15 phase numbers, compositions, as well as the temperature and pressure are determined at the maximum  
16 entropy state of system. (Qiu et al., 2014) has developed a consistent and efficient phase equilibrium-  
17 solver using the PR EoS based on the Lagrange-Eulerian framework of KIVA-3 (Amsden, 1997).  
18 Besides, there are many other well developed flash approaches, including the isochoric-isothermal  
19 (TVn) flash (Espósito et al., 2000), negative flash (Whitson and Michelsen, 1989) and isoenergetic-  
20 isochoric (UVn) flash (Castier, 2009). Among them, the UVn flash is an efficient tool for the phase  
21 change computations. Following this flash, the equilibrium pressure, temperature and phase  
22 compositions can be determined at the given specified mixture internal energy, volume and mole  
23 number of each component. Thereby, the UVn flash is identified as the thermodynamic method that  
24 should be applied in real-fluid solvers for the phase change computation under the thermodynamic  
25 equilibrium condition.

26

27 In this study, a detailed multicomponent real-fluid fully compressible model is presented, and the real-  
28 fluid wave patterns for water-nitrogen and *n*-dodecane/nitrogen mixtures are unraveled. The structure  
29 of this paper is as follows: Section 2 introduces the governing equations, a composite EoS, the  
30 numerical method, the hyperbolicity of two-phase Euler system, as well as the primary calculation  
31 steps. Section 3 provides the comparisons between the present model predictions and other available  
32 numerical results (Chiapolino et al., 2017), as well as the experimental measurements in flash boiling  
33 cases (Simões-Moreira and Shepherd, 1999). In Section 4, a series of shock and double-expansion  
34 tube cases for *n*-dodecane/nitrogen mixture are simulated, and the wave evolutions are unraveled.  
35 Section 5 presents a three-dimensional transcritical fuel injection case to highlight the performance of  
36 the proposed four-equation model. Finally, the conclusions are summarized in Section 6.

## 1 2. Numerical models

2 This paper presents a multicomponent two-phase flow model with real-fluid equilibrium-solver, which  
 3 is designed to be used for Eulerian large-eddy simulations (LES) of industrial two-phase flow  
 4 configurations (e.g., liquid-fuel injection in internal combustion engines). The model descriptions are  
 5 organized with the following three aspects. First, we introduce the governing equations of this model.  
 6 Then, the numerical methods are described, separately identifying the equations and unknowns in the  
 7 flow- and equilibrium-solvers, and highlighting the coupling between them. Finally, the hyperbolicity  
 8 of Euler system closed by the composite EoS for two-phase mixture is discussed.

9

### 10 2.1 Governing equations

11 In the two-phase flow system, one cannot fundamentally consider the thermal equilibrium at liquid-gas  
 12 interface in absence of thermal diffusion (Saurel et al., 2016), therefore, the Navier-Stokes equation is  
 13 more appropriate for two-phase flow systems. The starting point of this work is a filtered fully  
 14 compressible multicomponent Navier-Stokes equations, including the balance equations for distinct  
 15 species in gas and liquid phases, mixture momentum, and mixture specific internal energy as follows:

$$16 \quad \frac{\partial \alpha_g \rho_{k,g}}{\partial t} + \frac{\partial \alpha_g \rho_{k,g} u_i}{\partial x_i} = \dot{m}_k \quad (1)$$

$$17 \quad \frac{\partial \alpha_l \rho_{k,l}}{\partial t} + \frac{\partial \alpha_l \rho_{k,l} u_i}{\partial x_i} = -\dot{m}_k \quad (2)$$

$$18 \quad \frac{\partial \rho u_i}{\partial t} + \frac{\partial \rho u_i u_j}{\partial x_j} = \frac{\partial P}{\partial x_i} + \frac{\partial \tau_{ij}^{LT}}{\partial x_j} \quad (3)$$

$$19 \quad \frac{\partial \rho e}{\partial t} + \frac{\partial \rho e u_j}{\partial x_j} = -P \frac{\partial u_j}{\partial x_j} - \frac{\partial q^{LT}}{\partial x_j} + \tau_{ij}^{LT} \frac{\partial u_i}{\partial x_j} \quad (4)$$

20 As long as the multicomponent mixture is outside the vapor dome (i.e., single phase), the above  
 21 system is closed by PR EoS (Eqs. (5)). However, if the mixture is inside the vapor dome (i.e., two  
 22 phase), the system is closed by the composite EoS connected with the set of algebraic equations (Eqs.  
 23 (5) and (6)). This thermodynamic closing is inspired and very similar to the one used in the two-phase  
 24 flow models of (Saurel et al., 2008), (Petitpas et al., 2009), (Wareing et al., 2013), and (Saurel et al.,  
 25 2016), except for different EoSs used for each phase. In addition, in the composite EoS, each phase  
 26 always follows its own PR EoS, and the equilibrium connection constraints (Eqs. (6)) ensure that the  
 27 mixture speed of sound is always defined.

$$28 \quad \left\{ \begin{array}{l} P = \frac{RT}{V_p - b_p} - \frac{a_p \alpha_T}{V_p(V_p + b_p) + b_p(V_p - b_p)}, (p = 1: \text{gas}; p = 2: \text{liquid}) \\ C_{s,p}^2 = \frac{\partial P}{\partial \rho_p} \Big|_s = \frac{\partial P}{\partial \rho_p} \Big|_T + \frac{T}{c_{v,p} \rho_p^2} \left( \frac{\partial P}{\partial T} \Big|_{\rho_p} \right)^2 \end{array} \right. \quad (5)$$

$$29 \quad \left\{ \begin{array}{l} \rho = \sum \alpha_p \rho_p \\ e = \frac{1}{\rho} \sum \alpha_p \rho_p e_p \\ \frac{1}{\rho C_{s,mix,W}^2} = \sum \frac{\alpha_p}{\rho_p C_{s,p}^2} \end{array} \right. \quad (6)$$

1 where  $\rho_{k,p}$  denotes the partial density of species  $k$  in phase  $p$ ,  $p = 1$  for gas (index  $g$ ) and  $p = 2$  for  
2 liquid (index  $l$ );  $k \in [1, N]$ , and  $N$  is total species number;  $\alpha_p$  (with the saturation constant  $\sum \alpha_p = 1$ ),  
3  $\rho_p$ ,  $e_p$  and  $C_{s,p}$  are the phase volume fraction, density, specific internal energy, and speed of sound,  
4 respectively;  $\rho$ ,  $e$ ,  $P$ ,  $T$ ,  $u$  and  $C_{s,mix,W}$  are the mixture density, specific internal energy, pressure,  
5 temperature, velocity and Wood speed of sound (Wood, 1930), respectively (i.e., possible slip velocity  
6 between gas and liquid phases is neglected);  $R$  is the universal gas constant;  $\dot{m}_k$  is the phase change  
7 mass rate of species  $k$ , which will be determined by the isoenergetic-isochoic flash, described in  
8 Appendix A.3. In this model, the species diffusion flux is not considered, and the heat conduction flux  
9 is calculated based on the Fourier's law as  $q_j^{LT} = -\lambda^{LT} \nabla T$ ;  $\lambda^{LT}$  is the heat conduction coefficient  
10 covering laminar and turbulent contributions, written as  $\lambda^{LT} = \lambda^L + K^T \lambda^T$ , where  $K^T = 1$  for  
11 turbulent flows. The laminar contribution  $\lambda^L$  is computed by (Chung et al., 1988) correlation, and the  
12 turbulent one is estimated using a given turbulent Prandtl number,  $Pr_t = 0.9$ . The viscous stress tensor  
13 is composed of laminar and turbulent contributions and written as  $\tau_{ij}^{LT} = \tau_{ij}^L + K^T \tau_{ij}^T$ . As described in  
14 our previous work (Habchi, 2015), a Boussinesq approximation is used to compute  $\tau_{ij}^{LT}$ , for which a  
15 subgrid-scale turbulent viscosity given by Smogorinsky LES model is adopted, and the laminar  
16 viscosity is computed from (Chung et al., 1988) correlation.

17  
18 In the above system ((1) – (6)), there are three related volumes, including the mixture molar volume,  
19 phase molar volume, and partial volume ( $V, V_p, V_{k,p}$ ). The mixture molar volume is determined by the  
20 phase volume as  $V = \sum \varphi_p V_p$ , where  $\varphi_p$  is the phase molar fraction. Since a multicomponent mixture  
21 is considered in each phase,  $V_p$  is estimated based on the van der Waals mixing rule considering the  
22 interaction between molecular volume, energy and components. For subcritical two-phase conditions,  
23 the PR EoS is first solved based on the liquid phase compositions ( $x_k$ ) and three roots can be obtained.  
24 The smallest positive one is selected as the liquid molar volume ( $V_l$ ). Then, the PR EoS is solved  
25 based on the gas phase compositions ( $y_k$ ) and the largest one is selected as the gas molar volume ( $V_g$ ).  
26 For single-phase conditions, the PR EoS is solved based on the feed, and only the real positive molar  
27 volume is considered. When the phase molar volume is known, the phase density and mixture density  
28 are separately calculated by  $\rho_p = M_p/V_p$  and  $\rho = \sum \alpha_p \rho_p$ . Then, the partial density ( $\rho_{k,p}$ ) in Eqs. (1) –  
29 (6) is related to the phase density ( $\rho_p$ ) as  $\rho_{k,g} = \rho_g (y_k M_k / \sum y_k M_k)$  and  $\rho_{k,l} = \rho_l (x_k M_k / \sum x_k M_k)$ ,  
30 where  $M_k$  is the molecular weight of species  $k$ .

31  
32 Since the PR EoS has been demonstrated to underestimate the liquid hydrocarbon density, a volume  
33 translation method ( $V = V + V_{correct}$ ) has been used for improving the prediction of liquid density  
34 like (Baled et al., 2012, Tapriyal et al., 2012). The volume correction ( $V_{correct}$ ) is defined as:

$$V_{correct} = A + BT_r \quad (7)$$



1 where  $T_r$  is the reduced temperature ( $T_r = T/T_c$ );  $A$  and  $B$  are coefficients correlating the molecular  
2 weight ( $M$ ) and acentric factor ( $\omega$ ), formulated as:

$$3 \quad A, B = k_0 + k_1 \exp\left(\frac{-1}{k_2 M \omega}\right) + k_3 \exp\left(\frac{-1}{k_4 M \omega}\right) + k_5 \exp\left(\frac{-1}{k_6 M \omega}\right) \quad (8)$$

4 where  $k_1, k_2, k_3, k_4, k_5$  and  $k_6$  are the EoS based constants.

5  
6 It is worth noting that  $\alpha_p \rho_{k,p}$  is transported in our four-equation model, this is different from (Matheis  
7 and Hickel, 2018) model, in which partial densities of species ( $\rho Y_k$ ) in mixture are transported, where  
8  $Y_k$  is the mass fraction of species  $k$ . Indeed, the present formulation possesses some advantages, for  
9 instance, the knowledge of phase compositions obtained from the flow-solver can be used as  
10 initializations for UVn flash, as explained below in Section 2.2.

## 11 12 2.2 Numerical methods

13 This four-equation model has been implemented into an in-house IFP-C3D code (Bohbot et al., 2009)  
14 already including a seven-equation model with SG EoS (Habchi, 2015). IFP-C3D is an unstructured  
15 parallel solver with a finite volume formalism on staggered grids. As known, Eqs. (1) – (4) are the  
16 fully compressible Navier-Stokes equations with a parabolic nature. Without conduction and diffusion  
17 terms, Eqs. (1) – (4) are the Euler system (hyperbolic part). In order to solve Navier-Stokes equations,  
18 a fractional step approach is used, which is similar to the one used in (Wang et al., 2014). First,  
19 assume no phase change and the right hand side of species transport equations is set as zero, and only  
20 solve the flow-solver (Eqs. (1) – (4)) without  $\dot{m}_k$  terms. Then, an isolated system without flows into  
21 and out of the control volume is assumed, only consider the phase change and solve the phase  
22 equilibrium-solver (Eqs. (11) – (14)).

23  
24 In the flow-solver (Eqs. (1) – (4) without  $\dot{m}_k$ ), The parabolic and hyperbolic parts are solved  
25 separately and consecutively based on the time-splitting method in IFP-C3D (Bohbot et al., 2009). The  
26 time-splitting begins with an implicit Lagrangian stage, then follows a sub-cycled explicit Eulerian  
27 stage. In the Lagrangian stage, a second order implicit differencing is used for parabolic terms in Eqs.  
28 (1) – (4). The coupled implicit equations (velocity, pressure and temperature) are solved by SIMPLE  
29 algorithm (Patankar, 1980). Then, the obtained solutions are updated by solving the hyperbolic part in  
30 the Eulerian stage using a quasi-second-order-upwind (QSOU) explicit numerical scheme. The  
31 Minmod slope limiter is used for scalar fluxes, and the Van Leer slope limiter is used for momentum  
32 fluxes. In order to make sure the stability of using different TVD (total variation diminishing) schemes  
33 for momentum and scalars, simple advection of isolated interface cases have been tested. The results  
34 indicate that the combined method with Minmod and Van Leer limiters can get stable results with  
35 small oscillations, which are close to the results predicted by the same scheme. Considering the length  
36 of article, the compared results are not shown here. The above algorithms used in this study are known

1 to be able to ensure a stable quasi-second-order upwind advection for both scalars and momentum  
 2 (Bohbot et al., 2009).

3  
 4 Since this system is closed by the thermodynamic closure as Eqs. (5) – (6), it can yield (2N+7)  
 5 equations, including 2N species, one momentum, one energy balance equations, two PR EoS, three  
 6 connection constraints. Indeed, there are (2N+7) unknowns that need to be solved, including  $\rho_{k,g}$ ,  $\rho_{k,l}$ ,  
 7  $\alpha_g$  (and  $\alpha_l = 1 - \alpha_g$ ),  $e_g$ ,  $e_l$ ,  $u$ ,  $P$ ,  $\rho$  and  $e$ . After the flow-solver, we can get the equilibrium mixture  
 8 internal energy and density ( $e^*$ ,  $\rho^*$ ) denoted by (\*), as well as the temporary non-equilibrium variables  
 9 ( $\alpha''_g$ ,  $\alpha''_l$ ,  $\rho''_{k,g}$ ,  $\rho''_{k,l}$ ,  $e''_g$ ,  $e''_l$ ,  $P''$ ), denoted by ("), from which the temporary non-equilibrium phase  
 10 compositions can be obtained as  $x''_k = \alpha''_l \rho''_{k,l} M_k / \sum \alpha''_l \rho''_{k,l} M_k$  and  $y''_k = \alpha''_g \rho''_{k,g} M_k / \sum \alpha''_g \rho''_{k,g} M_k$ .

11  
 12 Basically, the final equilibrium temperature, pressure, and phase compositions ( $T, P, x_k, y_k$ ) will be  
 13 computed in the following phase equilibrium-solver. Then, the phase change mass ( $\dot{m}_k$ ) can be  
 14 obtained, and the volume fraction and partial densities will be updated as follows:

$$15 \quad \frac{\partial \alpha_g \rho_{k,g}}{\partial t} = \dot{m}_k \quad (9)$$

$$16 \quad \frac{\partial \alpha_l \rho_{k,l}}{\partial t} = -\dot{m}_k \quad (10)$$

17  
 18 The phase equilibrium-solver is mainly composed of an isochoric-isoenergetic (UVn) flash, and the  
 19 target variables are  $T, P, x_k, y_k$ . However, since the phase compositions ( $x_k, y_k$ ) are not independent  
 20 variables, the equilibrium-factors and vapor fraction ( $K_k, \psi_g$ ) are used instead. As long as  $K_k$  and  $\psi_g$   
 21 are known, the phase compositions ( $x_k, y_k$ ) can be obtained as  $x_k = z_k^* / (1 + (K_k - 1)\psi_g)$  and  
 22  $y_k = z_k^* K_k / (1 + (K_k - 1)\psi_g)$ , where  $z_k^*$  is the feed determined by inputs. Thus, the number of  
 23 unknowns in the phase equilibrium-solver is (N+3), including  $T, P, \psi_g$  and  $K_k$  ( $k = 1, N$ ). They are  
 24 subjected to the following set of (N+3) algebraic constraints:

$$25 \quad e^* = [\psi_g (M_g e_g) + (1 - \psi_g) (M_l e_l)] / \bar{M} \quad (11)$$

$$26 \quad \rho^* = \bar{M} / [\psi_g (M_g / \rho_g) + (1 - \psi_g) (M_l / \rho_l)] \quad (12)$$

$$27 \quad f_{k,l} / f_{k,g} = 1 \quad (13)$$

$$28 \quad \sum_{k=1}^N (z_k^* (1 - K_k) / (1 + (K_k - 1)\psi_g)) = 0 \quad (14)$$

29 where  $f_{k,p}$  is the fugacity of species  $k$  in phase  $p$ ;  $\bar{M}$  is the mixture molecular weight determined by the  
 30 feed; the liquid and gas internal energy, density and molecular weight ( $e_p, \rho_p, M_p$ ) are related to  
 31  $T, P, \psi_g$ . Clearly, based on the above constraints and initializations provided by the flow-solver, the  
 32 phase equilibrium-solver can be solved iteratively, using the composite EoS approach. In fact, the  
 33 constraints of Eqs. (11) and (12) are the same with those of  $e$  and  $\rho$  in Eqs. (6). Eq. (13) expresses the  
 34 equality of chemical potentials, as explained in Appendix A. 3, and the Eq. (14) is the well-known

1 Rachford-Rice equation which is an additional constraint to the equilibrium-solver as used in (Saha  
2 and Carrolls, 1997).

3

4 Indeed, the equilibrium-solver is composed of three parts:

5 (1) Phase stability test: it is performed by the Tangent Plane Distance (TPD) criterion (Michelsen,  
6 1982) to test the phase stability. The objective function is Eq. (A. 9), and the inputs include  
7 temperature, pressure, feed in the assumed phase ( $T, P, z_k$ );

8 (2) Isothermal-isobaric (TPn) flash: it is applied to obtain the equilibrium phase compositions ( $x_k, y_k$ ),  
9 and the objective functions are Eqs. (A. 13) – (A. 14). The inputs are temperature, pressure, feed,  
10 initial vapor fraction and equilibrium factors ( $T, P, z_k, \psi_g'', K_k''$ );

11 (3) Isoenergetic-Isochoric (UVn) flash: it is used to get the equilibrium temperature and pressure ( $T, P$ )  
12 considering the phase change, and the objective functions are Eqs. (A. 17) – (A. 18). The inputs  
13 include the mixture specific internal energy, density, feed, vapor fraction, as well as the initial  
14 temperature and pressure ( $e^*, \rho^*, z_k, \psi_g, T'', P''$ ).

15 The detailed phase equilibrium-solver is described in Appendix A.2 – A.3 to make the whole system  
16 clear and simplified.

17

18 For this compressible Navier-Stokes equations, the time-steps  $\Delta t_L$  (lagrangian stage) and  $\Delta t_E$  (Euler  
19 stage) are selected automatically at the beginning of each cycle. First, because the convective terms are  
20 explicitly sub-cycled, the convection time-step ( $\Delta t_E$ ) must satisfy the Courant stability condition.

21 Second, because the parabolic terms are implicitly discretized, there are no stability restrictions, but  
22 two temporal accuracy conditions, used for the selection of  $\Delta t_L$  in the implicit Lagrangian step. The  
23 first accuracy condition is related to the velocity gradient expressed as  $\Delta t_L \leq f_a \Delta x / (u^B - u)$ , where  
24  $f_a$  is the positive real number of order unity. The second accuracy condition is given in terms of the

25 strain tensor rate and calculated as  $\Delta t_L \leq f_r / \left( 2 \sqrt{\frac{1}{3}(p^2 - 3q)/3} \right)$ , where  $p$  and  $q$  can be found in

26 (Amsden et al., 1989). The convection time-step  $\Delta t_E$  must satisfy the Courant-Friedrichs-Lewy (CFL)  
27 condition ( $\Delta t_E \leq C_{CFL} \Delta x / (u + C_{s,mix})$ ) for the stability as the parabolic part of Navier-stokes  
28 equation is solved implicitly. Therefore, to ensure these conditions, the final time-step needs to be

$$29 \quad \Delta t = \min \left( C_{CFL} \Delta x / (u + c), f_a \Delta x / (u^B - u), f_r / 2 \sqrt{\frac{1}{3}(p^2 - 3q)/3} \right) \quad (15)$$

30

### 31 2.3 Hyperbolicity of Euler system with PR-EoS

32 Without considering the conduction, diffusion, and phase change terms, the system Eqs. (1) – (4) can  
33 be written in the compact form with corresponding initial conditions when dealing with the Riemann  
34 problem as

$$\begin{cases} \frac{\partial U}{\partial t} + \frac{\partial F(U)}{\partial x} = 0 \\ U(x, 0) = \begin{cases} U_L & \text{if } x \leq 0.5 \\ U_R & \text{if } x > 0.5 \end{cases} \end{cases} \quad (16)$$

where  $U = [\alpha_g \rho_{k,g}, \alpha_l \rho_{k,l}, \rho u, \rho e]^t$  are dependent variables, and  $F(U) = [\alpha_g \rho_{k,g} u, \alpha_l \rho_{k,l} u, \rho u^2 + P, (\rho e + P)u]^t$  presents the associated fluxes;  $U_L$  and  $U_R$  are the constant initial states in the left (L) and right (R) sides with the discontinuity located at  $x = 0.5$ . As long as all eigenvalues are real with eigenvectors linearly independent, the system (16) is hyperbolic with  $2N+2$  (only three distinct) eigenvalues:  $u - C_{s,mix}$ ,  $u$  ( $2N$ -folds) and  $u + C_{s,mix}$ , where  $C_{s,mix}$  is the mixture sound speed (Ma et al., 2017, Menikoff and Plohr, 1989, Petitpas et al., 2009, Saurel et al., 2016). Our Euler system (16) differs only in two aspects from the hyperbolic systems of (Saurel et al., 2016) and (Chiapolino et al., 2017). The main differences appear in: (1) we consider multicomponent in both liquid and gas phases, and (2) we use a composite PR EoS instead of Stiffened Gas EoS.

11

12 1) It is well known that our suggested model thermodynamically closed by a cubic equation of state  
 13 (e.g., PR EoS) is hyperbolic as long as the sound speed is real outside the vapor dome (see (Ma et  
 14 al., 2017), Appendix B). Here, the vapor dome denotes the region bounded by the bubble curve on  
 15 left and the dew curve on right, as shown in Fig. 1.

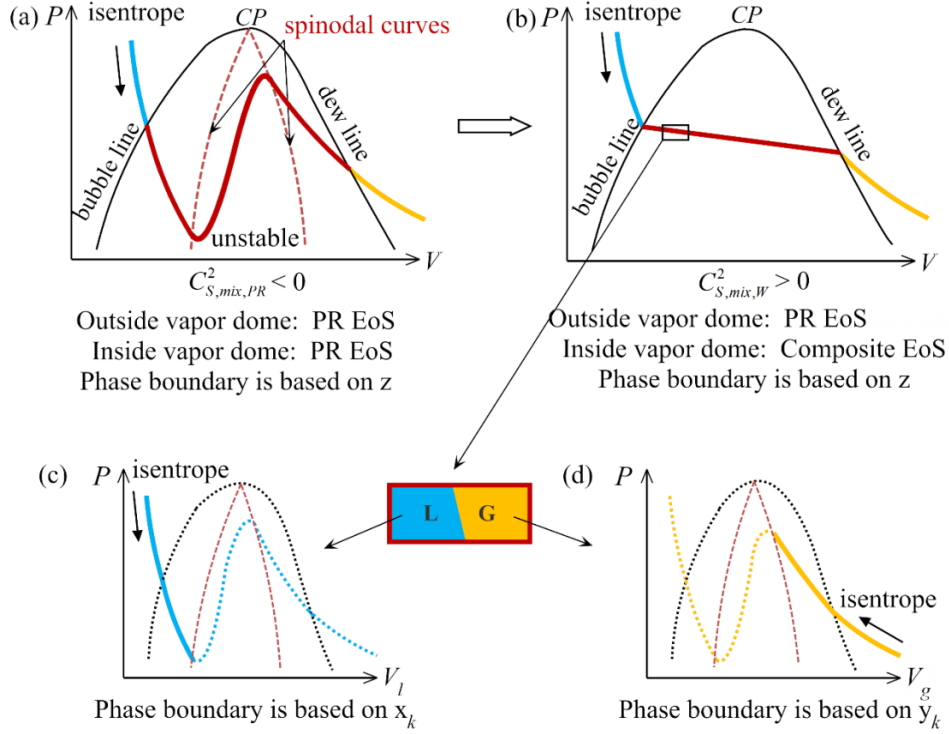
16

17 2) However, inside the vapor dome (i.e., two-phase), the issue is complex as the thermodynamic state  
 18 described by the ‘‘mixture PR EoS’’ is either metastable or unstable/non-convex (see Fig. 1(a)).  
 19 Here, the ‘‘mixture PR EoS’’ is defined as using the classical PR EoS for two-phase mixtures.  
 20 Inside the unstable/non-convex region bounded by the spinodal curves  $((\partial P / \partial v)_T = 0)$ , see Fig.  
 21 1(a)), there is a complex speed of sound (SoS) region where the hyperbolicity of Euler system is  
 22 lost, as depicted by Fig.27 in (Ma et al., 2017). It is therefore not appropriate to use the ‘‘mixture  
 23 PR EoS’’ to close the Euler system (16). Instead, a ‘‘composite EoS’’ formulation (see Eqs. (5) –  
 24 (6)) is used as the thermodynamic closure of the two-phase Euler system (16).

25

26 In our approach, each phase always has its own PR EoS, and the two-phase mixture state is obtained  
 27 by a ‘‘composite EoS’’ formulation. Each PR EoS is thermodynamically consistent (convex) with the  
 28 well-defined speed of sound (see Fig. 1(c) and (d)) outside its corresponding unstable region. The  
 29 connection between the two phases, being endowed by a ‘‘composite EoS’’, is carried out through a set  
 30 of algebraic constraints (Eqs. (6) are for the flow solver and Eqs. (11) – (14) are for the equilibrium  
 31 solver). Unlike the ‘‘mixture PR EoS’’ following a thermodynamic path (see Fig. 1(a)), the Wood  
 32 sound speed (Wood, 1930) used in the composite EoS only represents the mechanical equilibrium (see  
 33 Fig. 1(b)), and each phase separately follows its corresponding thermodynamic path as shown in Fig.  
 34 1(c) and (d) (Saurel et al., 2008). Due to  $C_{s,mix,PR}^2 \neq C_{s,mix,W}^2$ , no mixture thermodynamic path is

1 involved when solving the Euler system inside the vapor dome, the mixture speed of sound ( $C_{s,mix,W}$ )  
 2 is always defined. This is the reason why the present Euler system can preserve the hyperbolicity  
 3 inside the vapor dome (Saurel et al., 2008).  
 4



5  
 6 Fig. 1 Thermodynamic path along an isentrope for multicomponent flow. The vapor dome is enclosed  
 7 by the bubble and dew lines (a) mixture PR EoS: each phase follows its PR EoS, and the two-phase is  
 8 described by “mixture PR EoS”, the speed of sound is estimated as  $C_{s,mix,PR}^2 = (\partial P / \partial \rho)_s$ ,  $C_{s,mix,PR}^2$  is  
 9 negative in the unstable/non-convex region enclosed by spinodal curves ( $(\partial P / \partial V)_T = 0$ ), the phase  
 10 boundary is depicted based on the feed ( $z_k$ ); (b) a composite EoS: each phase has its own PR EoS, and  
 11 the two-phase mixtures are connected by a set of algebraic equations, in which the mixture sound  
 12 speed is calculated by the Wood formula as  $1 / (\rho C_{s,mix,W}^2) = \sum_p (\alpha_p / \rho_p C_p^2)$ ,  $C_{s,mix,W}^2$  is always  
 13 positive inside the vapor dome, the phase boundary is depicted based on the feed ( $z_k$ ); (c)  
 14 thermodynamic path along an isentrope for liquid inside the mixture, the phase boundary is depicted  
 15 based on liquid phase composition ( $x_k$ ),  $C_{s,l}^2 = (\partial P / \partial \rho_l)_s$ ; (d) thermodynamic path along an isentrope  
 16 for gas inside the mixture, the phase boundary is depicted based on gas phase composition ( $y_k$ ),  
 17  $C_{s,g}^2 = (\partial P / \partial \rho_g)_s$ .

18  
 19 Based on the studies of (Le Martelot et al., 2014, Lund, 2012, Saurel et al., 2008), it appears that the  
 20 mechanical equilibrium has the most significant impact on the sound speed, while the thermal and  
 21 chemical equilibrium assumptions have a much smaller effect. Therefore, there is no practical need to  
 22 compute the thermal and thermodynamic mixture speed of sound that is quite complex and

1 computationally expensive (Saurel et al., 2016). This is the reason why the Wood sound speed is  
 2 widely used although it has a little deviation from the thermodynamic equilibrium one, as shown by  
 3 (Le Martelot et al., 2014, Saurel et al., 2016, Saurel et al., 2008), to cite a few. Overall, the  
 4 consequence is that the composite EoS used in this study is a well compromised choice to keep the  
 5 suggested Euler system being hyperbolic, while using the PR EoS for each phase under an additional  
 6 numerical stability condition.

7  
 8 Indeed, since a fractional step approach is used to solve the Euler system, as described in Section 2.2,  
 9 we have met another issue in the composite EoS: the single phase inside the mixture may be  
 10 compressed or expanded due to the strong shock and rarefaction strength in the Lagrangian stage and  
 11 enter into the unstable region. To avoid the squared speed of sound of single phase ( $C_{s,p}^2$  given by Eqs.  
 12 (5)) becoming negative, a numerical stability criteria is added, in which the current cycle is restarted  
 13 with a smaller time-step ( $\Delta t$ ) when  $C_{s,p}^2 < 0$  ( $p = 1$ : gas,  $p = 2$ : liquid). Therefore, our model is able  
 14 to deal with metastable states in the lagrangian step of the Euler system solver. In fact, the additional  
 15 numerical stability criterion has been proved to intervene only in the cases involving very strong shock  
 16 and rarefaction waves (e.g., cavitation). Future work should develop better numerical scheme for the  
 17 Euler system, avoiding such fractional step approach.

#### 18 19 2.4 Calculation procedure

20 The four-equation model is solved by a fractional step approach (see Fig. 2), including the following  
 21 primary steps:

- 22 1) Read input parameters, including the temperature, pressure, and mass fraction in gas ( $T_0, P_0, Y_{g,0}$ );
- 23 2) Apply TPD function (see Appendix A.2) to test the phase stability;
- 24 3) If  $TPD = 0$  or  $1$ , the mixture is in single-phase (gas or liquid), and the initial compositions are set  
 25 as feed ( $z_k$ ). Otherwise, the flow is unstable and initial compositions of each phase ( $x_{k,0}, y_{k,0}$ ) are  
 26 determined by TPn flash (see Appendix A.3.1);
- 27 4) Determine the thermal and transport properties, including the conductivity, viscosity, and the  
 28 thermodynamic partial derivatives, described in the Appendix A.1;
- 29 5) In the flow-solver, assuming no phase change, only solve Eqs. (1) – (4) without  $\dot{m}_k$  term based on  
 30 the SIMPLE algorithm, QSOU scheme, using Van Leer and Minmod limiters. The composite EoS  
 31 (Eqs. (5) – (6)) is used to compute the thermodynamic properties;
- 32 6) After the flow-solver, we can get the mixture specific internal energy and density ( $e^*, \rho^*$ ), which  
 33 are constraints for the following phase equilibrium-solver (Eqs. (11) – (14)). The non-equilibrium  
 34 partial density in each phase ( $\rho''_{k,p}$ ) obtained from the flow-solver is used as the initialization for  
 35 the phase equilibrium-solver;

- 1 7) In the phase equilibrium-solver, assuming no flow and only considering the phase change, apply
- 2 the TPD function to check the phase stability;
- 3 8) If the mixture is in single-phase (TPD = 0 or 1), no phase change is assumed. The temperature and
- 4 pressure are iterated until the equilibrium state of  $e^*, \rho^*$  based on Eqs. (11) – (14) is obtained;
- 5 9) Otherwise, for the two-phase mixture (TPD = 2), Eqs. (11) – (14) are iteratively solved to
- 6 determine the equilibrium temperature and pressure ( $T, P$ ), as well as the phase compositions ( $x_k,$
- 7  $y_k$ ) considering the phase change, and the details are in Appendix A.3; It is important to note that
- 8 the sound speed is not needed during the iterative relaxation process.
- 9 10) At the end of each iteration of UVn flash, if the given tolerance is satisfied, go to the step (11)
- 10 directly, otherwise, the TPD test is applied to determine the phase stability again, and go to the
- 11 step (7) or (8) depending on the new TPD value;
- 12 11) Finally, the thermal and transport properties are updated with the equilibrium temperature,
- 13 pressure, and compositions;
- 14 12) Return to step (5) and repeat the above processes for the next time-step.

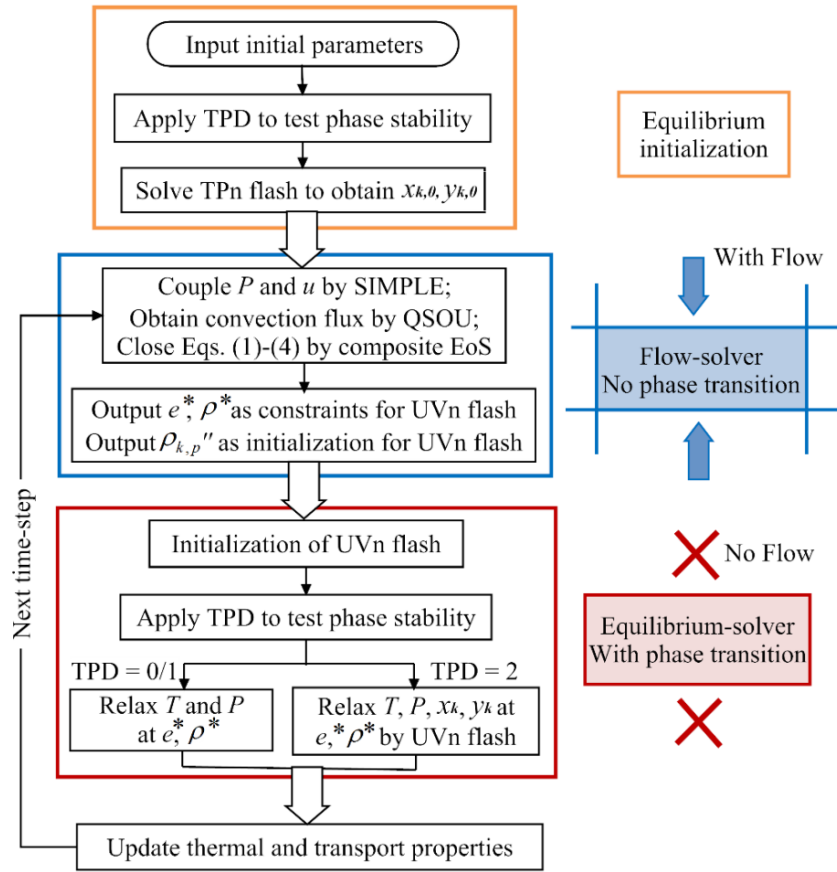


Fig. 2 Flowchart of the four-equation model with a fractional step approach.

### 3. Comparison and Validation

This four-equation model is applied to simulate the behaviors of multicomponent two-phase flow in the 1D shock and double-expansion tubes. In Section 3.1, the four-equation model predictions are

1 compared to the results of (Chiapolino et al., 2017) for shock and double-expansion tubes, followed by  
 2 Section 3.2, in which flash boiling cases are simulated and the model predictions are validated against  
 3 experimental measurements (Simões-Moreira and Shepherd, 1999). Section 3.3 illustrates the  
 4 sensitivity of mesh size, time-step, and convergence tolerance of UVn flash. All cases are simulated  
 5 using a 1 m long tube and a uniform 1D mesh.

### 6 3.1 Comparison with the available numerical results

7 To evaluate the correctness of implementation, two 1D cases (Table 1) for the water-air mixture  
 8 separately under shock and double-expansion tube conditions are firstly simulated, and compared with  
 9 the results of (Chiapolino et al., 2017) model to carry out a qualitative validation. (Chiapolino et al.,  
 10 2017) model is referred as 4EQ-SM-SG and the four-equation model in this study is termed as 4EQ-  
 11 MM-PR. For simplicity, the air is replaced by nitrogen ( $N_2$ ) in this study. In Chiapolino et al. (2017)  
 12 model, the flow system is similar to our four-equation model, both phases are compressible and  
 13 assumed in mechanical and thermodynamic equilibrium. However, there are two differences between  
 14 these two models. The first arises from the multicomponent nature (MM) of our transport equations in  
 15 liquid phase (Eq. (2)), while the dissolved gas in liquid phase is ignored in Chiapolino et al. (2017)  
 16 model. Second, the EoSs are different. The Noble Abel Stiffened Gas (NASG) EoS is used in  
 17 Chiapolino et al. (2017) model to close the governing equations and describe the thermal properties,  
 18 which have been validated with experimental data in a limited temperature and pressure ranges.  
 19 Therefore, their results are expected to be very close to the real fluids in these specified ranges, which  
 20 are valuable as references to verify our implementation qualitatively.

21  
 22  
 23 Table 1. Initial parameters for two-phase shock and double-expansion tube cases with water-nitrogen

	$P_L$ (MPa)	$P_R$ (MPa)	$T_L$ (K)	$T_R$ (K)	$u_L$ (m/s)	$u_R$ (m/s)	$\alpha_{g,L}$ Equilib.	$\alpha_{g,R}$ Equilib.	$Y_{H_2O,L} = Y_{H_2O,R}$ Input
Case 1	0.2	0.1	354	337	0.0	0.0	0.9995744	0.99977	0.3
Case 2	0.1	0.1	293	293	-1.0	1.0	0.9851587	0.98517	0.99998

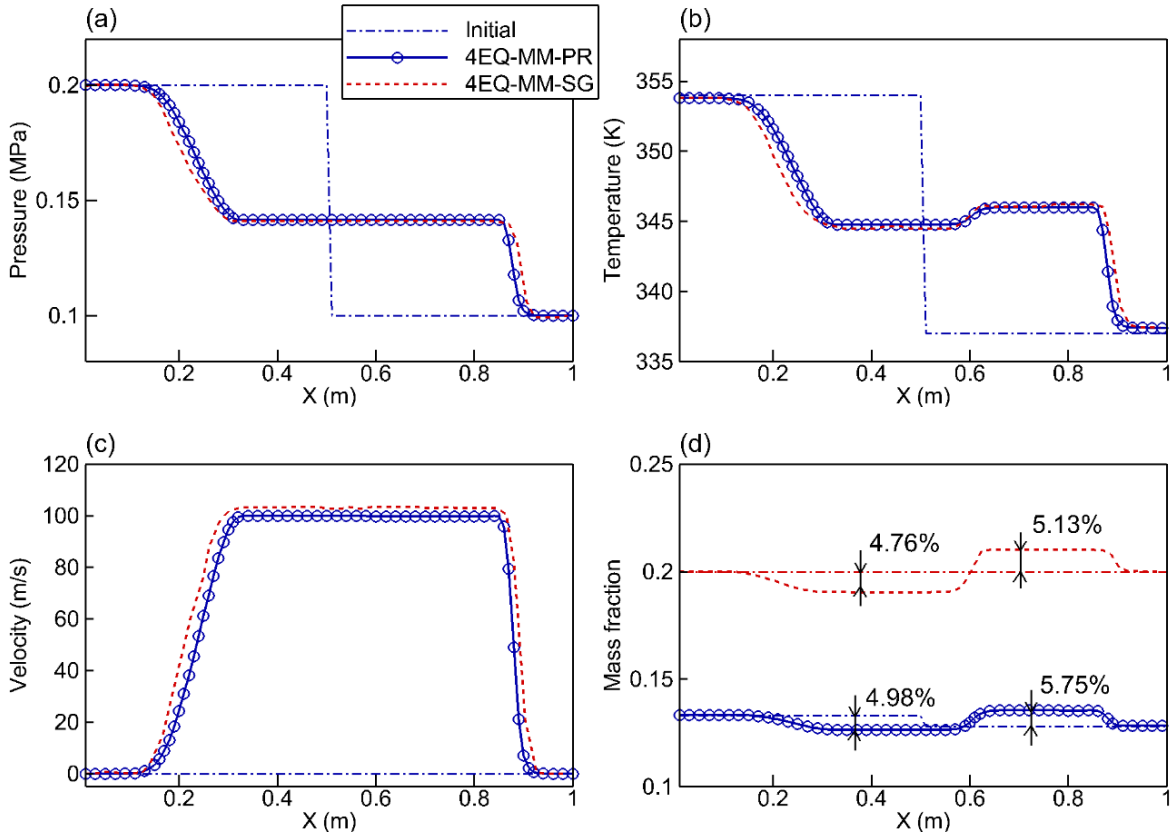
#### 24 3.1.1 Results of Case 1

25 First, a 1D shock tube is simulated (Case 1 in Table 1). The tube is filled with a homogeneous water-  
 26 nitrogen mixture, and the initial discontinuity is located at 0.5 m. The left side (0.2 MPa) is at a higher  
 27 pressure than the right side (0.1 MPa). The temperatures in both sides are set the same as those in  
 28 (Chiapolino et al., 2017). The compared results between the present four-equation and Chiapolino et al.  
 29 (2017) models at the instant of 1.0 ms are plotted in Fig. 3. It is found that good agreements were  
 30 obtained in terms of shock wave strength (velocity magnitude), as well as the evolution of pressure  
 31 and temperature at the contact discontinuity. However, there is a discrepancy in the vapor mass  
 32 fraction profile as shown in Fig. 3(d). This is because an isothermal-isobaric flash coupled with the PR  
 33



1 EoS is used in this study to estimate the equilibrium vapor mass fraction, which will cause a minor  
 2 difference with that of Chiapolino et al. (2017) model even though in the same initial temperature,  
 3 pressure, and feed. In addition, as the dissolved gas in liquid phase is neglected in Chiapolino et al.  
 4 (2017) model, it will also lead to a negligible deviation in the vapor mass fraction prediction. Even  
 5 though the vapor mass fraction predicted by these two models are different, their wave evolutions are  
 6 very similar, and the phase change proportion predicted by these two models are comparable. In this  
 7 section, we only want to carry out a qualitative comparison to validate our model implementation.  
 8 Therefore, the reasonable difference between two numerical results can be acceptable.

9



10

11

12 Fig. 3 Compared results between the four-equation (4EQ-MM-PR) and (Chiapolino et al., 2017)  
 13 (4EQ-MM-SG) models (Case 1:  $P_L = 0.2$  MPa,  $P_R = 0.1$  MPa,  $T_L = 354$  K,  $T_R = 337$  K,  $\alpha_{g,L} =$   
 14  $0.9995744$ ,  $\alpha_{g,R} = 0.9997698$ ,  $t = 1.0$  ms, thin dash dot lines are initials, 100 cells, CFL = 0.2).

15

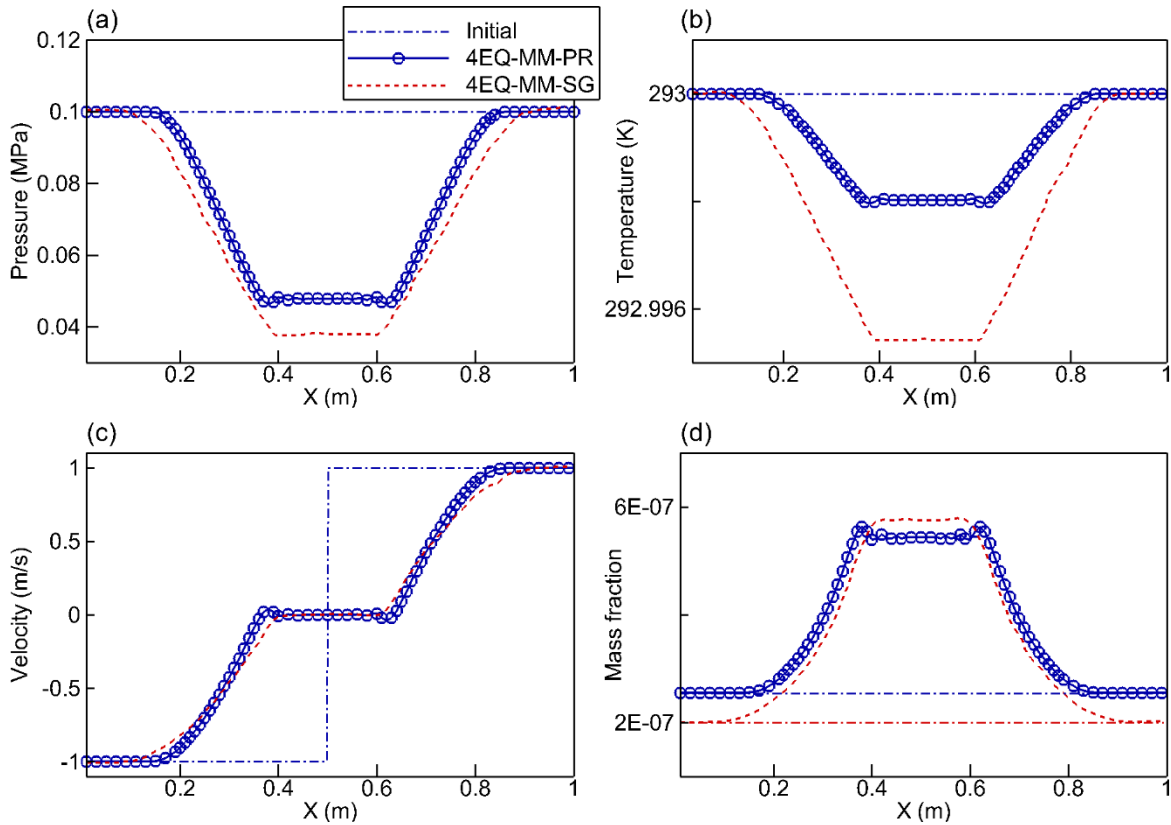
### 16 3.1.2 Results of Case 2

17 The double-expansion tube case also known as cavitation test is performed in a 1 m long tube filled  
 18 with water combined with a small volume fraction of gaseous nitrogen at atmospheric pressure. The  
 19 initial discontinuity is set at 0.5 m, and the left and right velocities are -1.0 m/s and 1.0 m/s,  
 20 respectively.

21

1 The expansion wave is generated towards to the left side combined with the evaporation front  
 2 progressing. The compared results between the present four-equation and (Chiapolino et al., 2017)  
 3 models are presented in Fig. 4. Significant discrepancies are observed in the pressure, temperature and  
 4 vapor mass fraction predictions between these two models. This is because the dissolved gas in liquid  
 5 phase is neglected in Chiapolino et al. (2017) model, the equilibrium pressure caused by the expansion  
 6 wave is equal to the vapor pressure of water. However, in the present four-equation model, the  
 7 dissolved nitrogen in liquid phase increases the mixture saturation pressure, which qualifies that the  
 8 final equilibrium pressure with current model is slightly higher than that in Chiapolino et al. (2017)  
 9 model. On the other hand, since the composite PR EoS is known to underestimate the water density,  
 10 the temperature is overestimated at the equilibrium state. A more detailed discussion on the influence  
 11 of liquid density on expansion behaviors is given in Section 4.2.

12



13

14

15 Fig. 4 Compared results between the four-equation (4EQ-MM-PR) and (Chiapolino et al., 2017)  
 16 (4EQ-SM-SG) models (Case 2:  $P_L = P_R = 0.1$  MPa,  $T_L = T_R = 293$  K,  $\alpha_{g,L} = \alpha_{g,R} = 0.9851587$ ,  
 17  $u_L = -1$  m/s,  $u_R = 1$  m/s,  $t = 3.5$  ms, thin dash dot lines are initials, 100 cells, CFL = 0.2).

18

### 19 3.2 Flash boiling test cases

20 In order to validate the accuracy of present four-equation model, several flash boiling test cases are  
 21 simulated and the results are compared with the experimental results (Simões-Moreira and Shepherd,  
 22 1999). In this flash boiling experiment, a high pressure tube filled with liquid *n*-dodecane at

1 thermodynamic equilibrium state is connected to a low pressure chamber with gaseous nitrogen, and  
 2 the detailed parameters of Case 3 are listed in Table 2. The flash boiling has been observed at the  
 3 liquid-gas interface in the experiment. It has been demonstrated that this kind of flash boiling have  
 4 similar behaviors with deflagration phenomena in combustion (Kurschat et al., 1992, Thompson et al.,  
 5 1987). An evaporation front propagates into the liquid side with a steady mean velocity,  $U_F$ , which has  
 6 been measured under various superheated conditions (Simões-Moreira and Shepherd, 1999). In this  
 7 study, the front velocity,  $U_F$ , is estimated based on a simple control volume model and Rankine-  
 8 Hugoniot equation (Simões-Moreira and Shepherd, 1999), as follows

$$9 \quad J = (U_F + u_L)\rho_L = (U_F + u_R)\rho_R \quad (17)$$

10 where  $u_L$  and  $u_R$  are the fluid velocity of left and right sides, respectively; and  $J$  is the superficial mass  
 11 flux. Since the liquid in left side is stationary ( $u_L = 0$ ), the front velocity can be calculated as

$$12 \quad U_F = J/\rho_L \quad (18)$$

$$13 \quad J^2 = (P_L - P_R)/(1/\rho_R - 1/\rho_L) \quad (19)$$

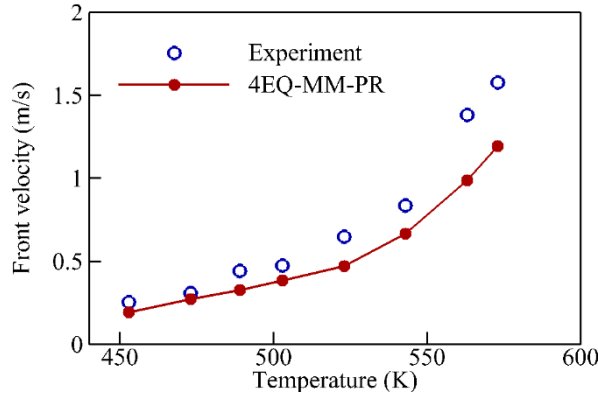
14 where  $P_L$  and  $\rho_L$ , as well as  $P_R$  and  $\rho_R$  refer to the pressure and density before and after the  
 15 evaporation front, respectively.

16  
 17 Table 2. Initial conditions for flash boiling cases with *n*-dodecane/nitrogen mixture

	$P_L$ (MPa)	$P_R$ (MPa)	$T_L$ (K)	$T_R$ (K)	$u_L = u_R$ (m/s)	$\alpha_{g,L}$ (Equilib.)	$\alpha_{g,R}$ (Equilib.)
Case 3	0.15, 0.22, 0.30, 0.39, 0.50, 0.75, 1.10, 1.30	0.0001	453, 473, 489, 503, 523, 543, 563, 573	543	0.0	0.0001	0.9999

18  
 19 The evaporation front velocities are obtained at several times in the range of 0.2 – 0.4 ms, and the  
 20 averaged values are compared with the experimental measurements as shown in Fig. 5. It is found that  
 21 the present model can obtain good agreements with experimental results under low superheated  
 22 conditions (i.e., less than 503 K). As the superheat degree rises, the discrepancy between experimental  
 23 results and model predictions increases. Here, the superheat degree is usually defined as the  
 24 temperature difference between the local temperature and the saturation temperature at a given  
 25 subcritical pressure. This may be caused by several reasons, including complexities and uncertainties  
 26 in experiment and assumptions used in the four-equation model. Indeed, unstable interfaces can be  
 27 observed in experimental images (Fig. 5 in (Simões-Moreira and Shepherd, 1999)) due to the high  
 28 nucleation rate near phase interface and also at walls. Some small droplets are busted from the  
 29 interface and flung into the flows, which enhances the gas velocity ( $u_R$ ). Following (17), the front  
 30 velocity can be described as  $U_F = u_R\rho_R/(\rho_L - \rho_R)$ . Due to the increased  $u_R$  caused by bursting  
 31 droplets observed in the experiment, the measured front velocity  $U_F$  is enhanced, and the increased  
 32 extent is growing as the superheat degree rises. Since, however, these effects are neglected in the

1 simulations, the present model underestimates the front velocity at the conditions with high superheat  
 2 degree. For instance, as the temperature of left side is 573 K, the front velocity is underestimated  
 3 around 25% as shown in Fig. 5. Therefore, the future work shall include the modelling of the amount  
 4 of droplets bursting at the interface. Indeed, this phenomenon is important for modeling of gasoline  
 5 direct injection (GDI) with flash boiling conditions.



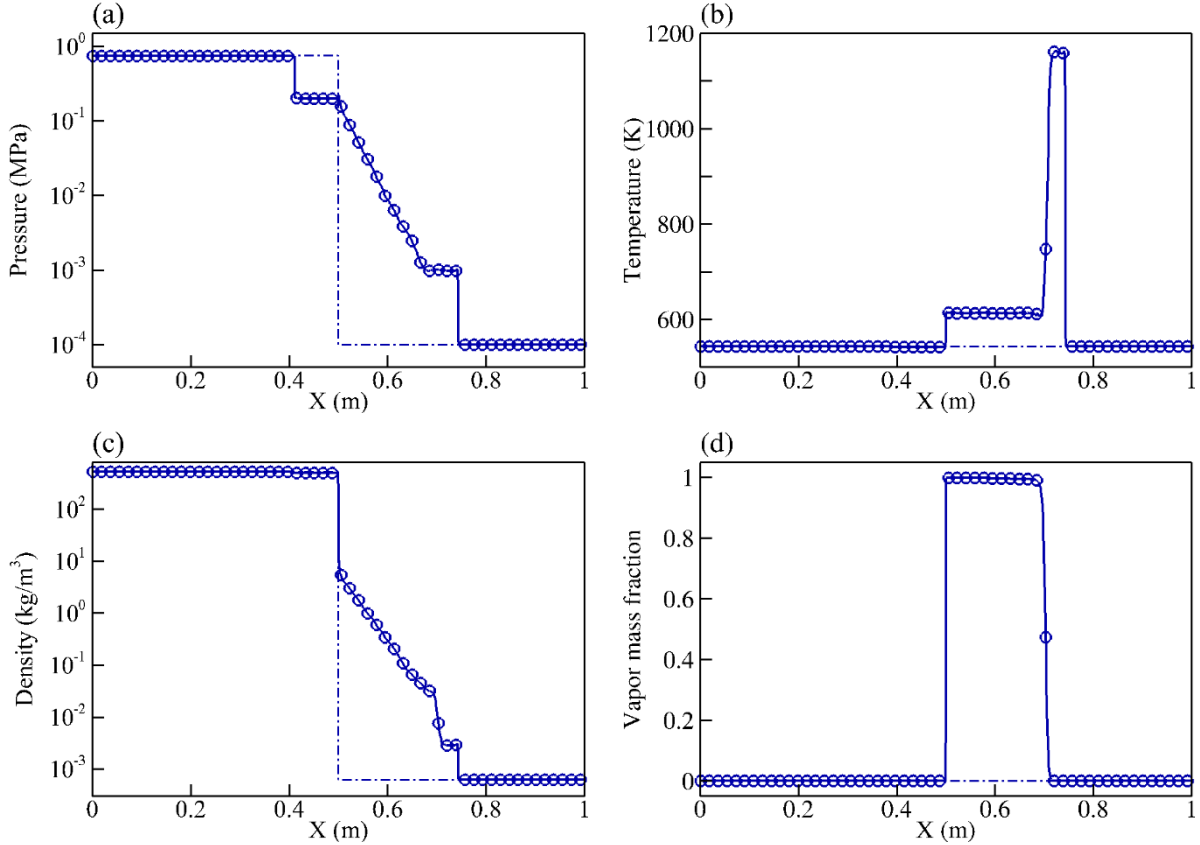
7  
 8 Fig. 5 Compared evaporation front velocity between experimental results (Simões-Moreira and  
 9 Shepherd, 1999) and the four-equation model predictions for flash boiling case (Case 3:  $P_L = 0.15 -$   
 10  $1.30$  MPa,  $P_R = 0.0001$  MPa,  $T_L = 453 - 573$  K,  $T_R = 543$  K,  $\alpha_{g,L} = 0.0001$ ,  $\alpha_{g,R} = 0.9999$ ,  $t =$   
 11  $0.2 - 0.4$  ms, left side is filled with liquid *n*-dodecane, right side is filled with gaseous nitrogen).

12  
 13 The evolution profiles of the flash boiling case with the liquid temperature of 543 K are presented in  
 14 Fig. 6. Since the evaporation in flash boiling cases takes place within a very thin region, its thickness  
 15 is quite small in comparison with the fluid length scales. Therefore, there is a sharp discontinuity at the  
 16 evaporation front ( $X = 0.5$  m) in each profile. In the left side, the pressure drops drastically across the  
 17 expansion wave, and the superheated liquid is transported along with the evaporation front to the left  
 18 side at a low speed. The vapor mass fraction increases suddenly to one at the liquid-gas interface.  
 19 Meanwhile, at the right side, a strong shock wave is produced and propagates towards the low-  
 20 pressure side. Between the evaporation and shock wave fronts, a simple contact discontinuity (in the  
 21 range of  $0.68 - 0.74$  m) is formed to connect the vapor front and shocked gas (Saurel et al., 2008).

22  
 23 The evolution of fluid velocity and mixture sound speed in different superheated cases are presented in  
 24 Fig. 7(a) and (b). In this figure, the labels A, B, and C are identified as the evaporated vapor mixture,  
 25 the contact discontinuity zone behind shock front, as well as the initial nitrogen gas zone, respectively.  
 26 It is found that the sonic velocity is reached very fast at the evaporation front ( $u = C_{s,mix}$ ), and then  
 27 the flow evolves to a supersonic regime. In this regime, the vapor velocity increases up to a constant  
 28 value (at zone B), which increases with raising the superheat degree (see Fig. 7(a)). It is noted that the  
 29 sound speed at zone A is very low ( $\sim 174.0$  m/s), which is consistent with NIST data for gaseous *n*-

1 dodecane (Linstrom and Mallard, 2001) at the prevailing temperature ( $\sim 600$  K) and pressure ( $\sim 1$  kPa),  
 2 as can be seen in Fig. 6(a) and (b). Since the fluid temperature at the contact discontinuity is very high  
 3 (see  $X \approx 0.68 - 0.74$  in Fig. 6(b)), the corresponding sound speed is very large (see zone B in Fig.  
 4 7(b)). However, both of them decrease suddenly at the shock front. The detailed wave behaviors are  
 5 further discussed in the following Section 4.1.2. Based on the above validated results and analysis, the  
 6 present four-equation model is demonstrated to be able to simulate the complex wave behaviors at the  
 7 interface with large discontinuities.

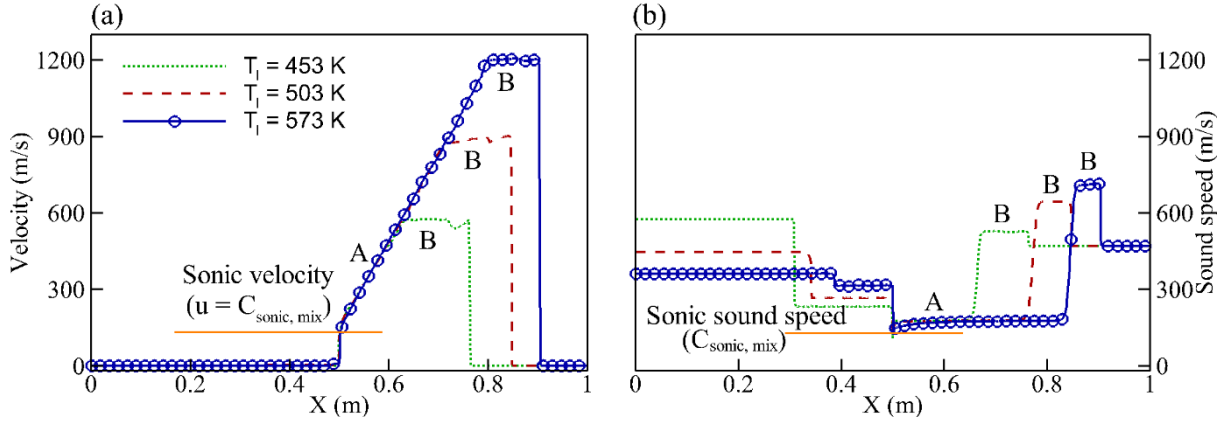
8



10

11 Fig. 6 Wave behaviors of flash boiling case predicted by the four-equation model ( $P_L = 0.75$  MPa,  
 12  $P_R = 0.0001$  MPa,  $T_L = 543$  K,  $T_R = 543$  K,  $\alpha_{g,L} = 0.0001$ ,  $\alpha_{g,R} = 0.9999$ ,  $t = 0.2$  ms, left side is  
 13 filled with liquid *n*-dodecane, right side is filled with gaseous nitrogen, thin dash dot lines are initials).

14



1  
2 Fig. 7 Fluid velocity and sound speed profiles of flash boiling case predicted by the four-equation  
3 model ( $P_L = 0.15$  MPa,  $T_L = 453$  K;  $P_L = 0.39$  MPa,  $T_L = 503$  K;  $P_L = 1.30$  MPa,  $T_L = 573$  K,  $\alpha_{g,L} =$   
4  $0.0001$ ,  $\alpha_{g,R} = 0.9999$ ,  $t = 0.3$  ms)

### 5 6 3.3 Numerical sensitivity analysis

7 The analysis of mesh size, time-step, and convergence sensitivity is a pre-requisite for the evaluation  
8 of the suggested four-equation model accuracy. Several cases have been simulated, and similar  
9 behaviors have been obtained. The results of three cases are presented in this section.

#### 10 11 3.3.1 Mesh size sensitivity analysis

12 In order to check the current model in dealing with numerical oscillations, a simple 1D advection of  
13 isolated interface case is simulated with Euler equations (Eqs. (1) – (4) without conduction, diffusion  
14 and phase change terms) inspired by the work of (Beig and Johnsen, 2015). The tube length is 1 m, the  
15 initial temperature and pressure of the whole domain ( $X \in [0, 1]$ ) filled with n-dodecane and nitrogen  
16 are 300 K and 1 bar, respectively. The isolated liquid zone ( $Y_{C_{12}H_{26},l} = 0.99$ ) ranging from 0.25 to  
17 0.75 m moves at a constant speed of 100 m/s to the right side. The gas zone is full of the mixture with  
18  $Y_{C_{12}H_{26},g} = 0.01$ . The numerical results depicted in Fig. 8 clearly indicate that our model produces  
19 some oscillations in pressure, temperature, and velocity, and these oscillations accumulate with wave  
20 propagating. As the mesh is refined, the oscillations are reduced, less than 0.01% with 2000 cells. This  
21 corroborates similar results with (Terashima and Koshi, 2012), for instance.

22  
23 Thereby, we can conclude that the present four-equation model can be used with confidence when the  
24 flow gradients are refined appropriately. In the future work, automatic mesh refinement (AMR) will be  
25 used with criteria based on density, mass fraction and heat capacity in order to minimize spurious  
26 oscillations. More sophisticated approaches like the one of (Pantano et al., 2017) coupled with the PR  
27 EoS may be another good option for our future studies.

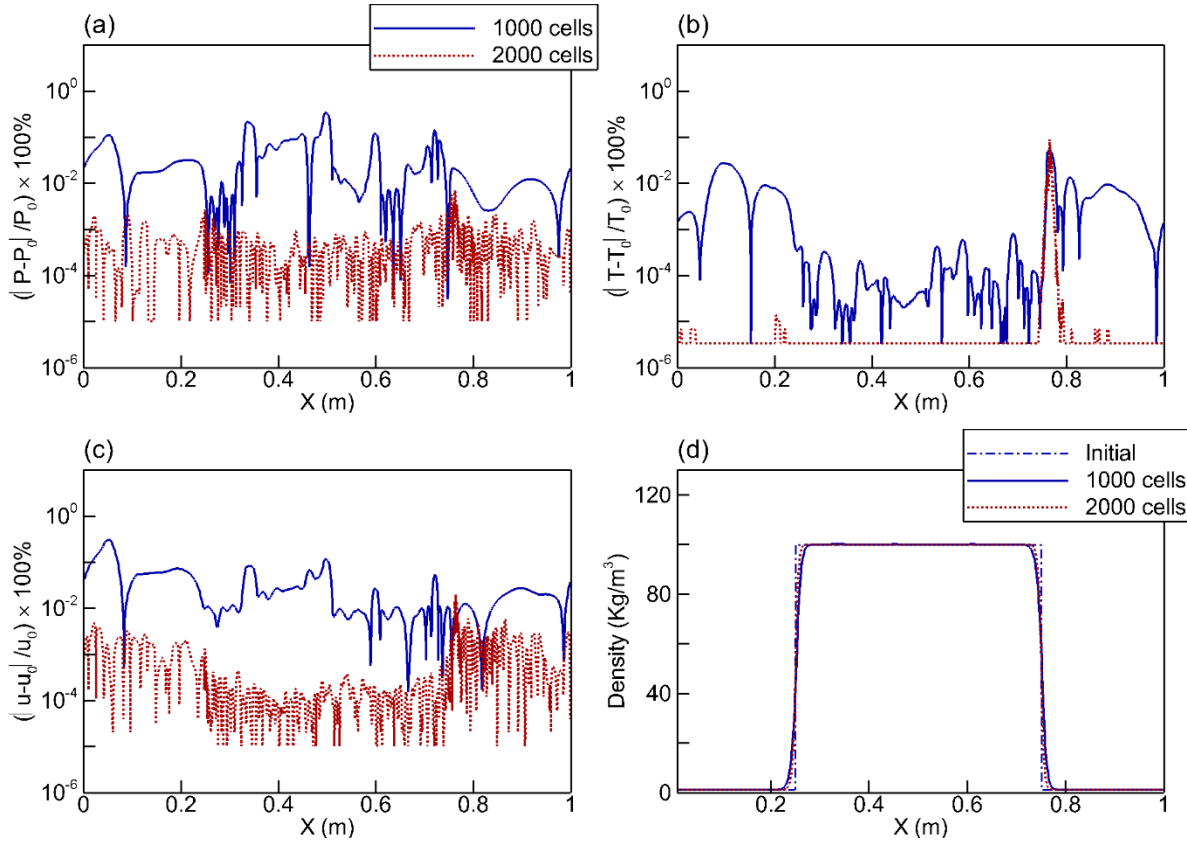


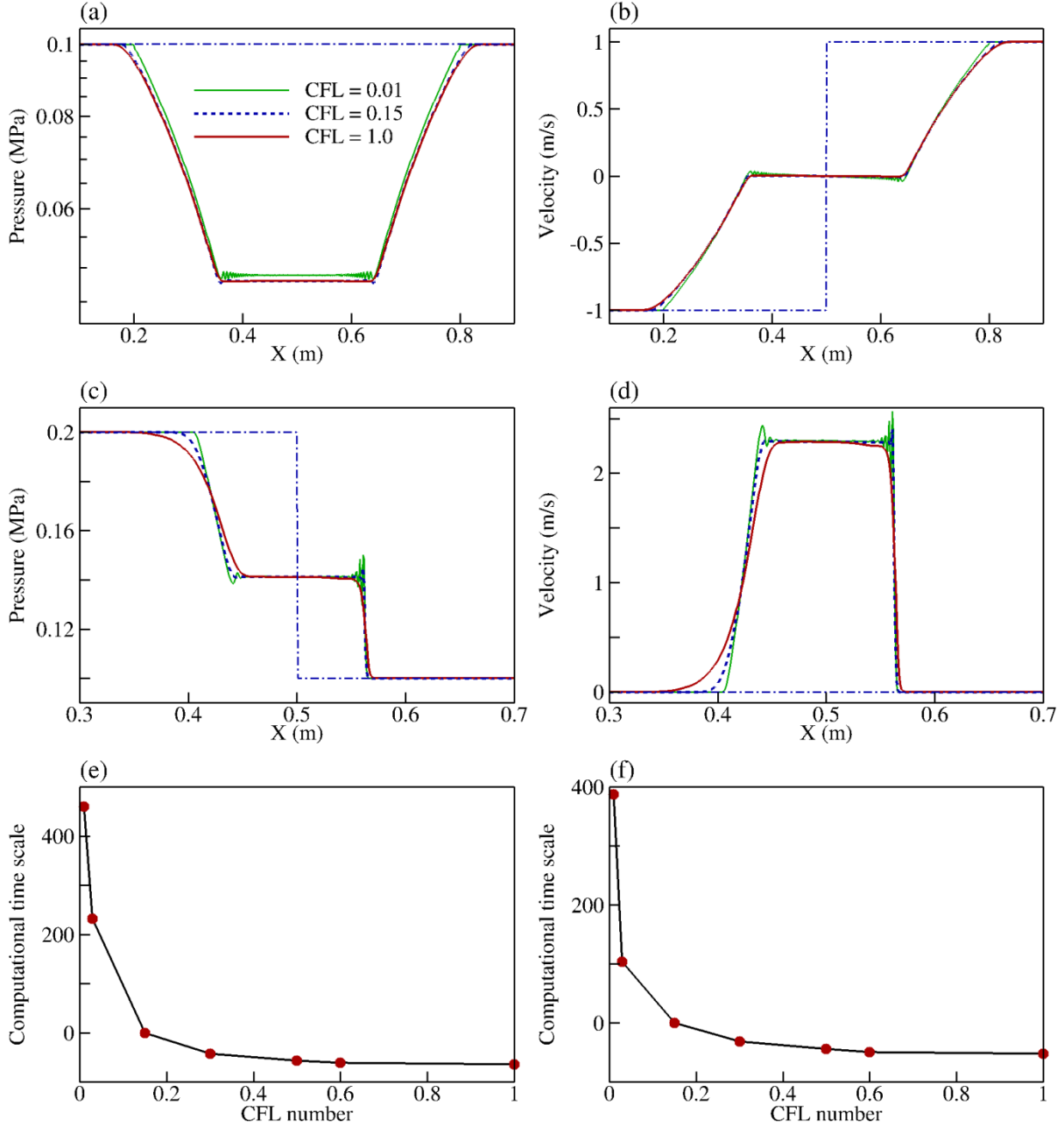
Fig. 8 Results of an isolated interface advection tube filled with n-dodecane/nitrogen mixture at 10 ms, errors in (a) pressure, (b) temperature, (c) velocity, and (d) density profile (thin blue dash dot is initial and exact solution, blue solid is 1000 cells, red dashed is 2000 cells,  $P = 1$  bar,  $T = 300$  K,  $Y_{C_{12}H_{26},l} = 0.99$ ,  $Y_{C_{12}H_{26},g} = 0.01$ ,  $u = 100$  m/s,  $CFL = 0.2$ ).

### 3.3.2 Time-step sensitivity analysis

Another primary numerical parameter is the time-step. The sensitivities of four-equation model to the CFL (Courant–Friedrichs–Lewy) number were investigated, and the results with CFL number in the range of 0.01 – 1.0 for a double-expansion tube (Case 2 in Section 3.1.3) and a shock tube (Case 4 in Section 4.1.1) are plotted in Fig. 9. Numerical instability on the solutions can be observed in the expanding cavitation interface in the case of smallest CFL number, and the instability amplitude is decreased as the CFL number increases. This is because in the present numerical solver, the parabolic terms are solved using the implicit central-differenced method, and hyperbolic is solved explicitly with a second-order upwind approach. Using small CFL number reduces the dissipation of dispersive waves. Therefore, the spurious oscillations increase in time since the dispersion and dissipation errors accumulate as the wave propagates. However, as a large CFL number is used, the numerical diffusion increases across the expansion and shock waves. As expected, the computational time decreases rapidly with raising the CFL number from 0.015 to 0.15. However, as the CFL number subsequently increases, the computational time show insignificant improvement in the computational efficiency. Consequently, based on these numerical results, the best CFL number for four-equation model is in the

1 range of 0.15 – 0.3 considering the computational efficiency and stability. In addition, it is noted that  
 2 the time-step is controlled by the CFL condition when  $C_{CFL}$  is small enough. On the contrary, as  $C_{CFL}$   
 3 is larger than 0.6, the time-step becomes rather controlled by the accuracy conditions.

4



5

6

7

8 Fig. 9 Time-step analysis (CFL number) for Case 2 (a, b, e,  $t = 3.5$  ms) and Case 4 (c, d, f,  $t = 2.0$  ms),  
 9  $\varepsilon_{UVn} = 1.0e-13$ , computational time scale =  $100 \times (t_{CFL} - t_{CFL=0.15})/t_{CFL=0.15}$

10

### 11 3.3.3 Convergence sensitivity analysis

12 To examine the behaviors of UVn flash solver, a convergence analysis was performed. The  
 13 convergence tolerance is termed as  $\varepsilon_{UVn}$ . All the objective functions involved in the UVn flash follow  
 14 the same tolerance. The convergence tolerance is defined as following:



1 
$$\left| \frac{\psi_g - \psi_{g,cal}}{\psi_g} \right| \leq \varepsilon_{UVn} \quad (20)$$

2 
$$\left( \ln \left( \frac{f_{k,l}}{f_{k,g}} \right) \right)^2 \leq \varepsilon_{UVn} \quad (21)$$

3 
$$\left( \frac{e^* - e_{cal}}{e^*} \right)^2 + \left( \frac{\rho^* - \rho_{cal}}{\rho^*} \right)^2 \leq \varepsilon_{UVn} \quad (22)$$

4 where the subscript *cal* indicates the calculated values during each iteration. The details of each  
5 parameter and objective functions in the UVn flash are introduced in Appendix A.3.

6

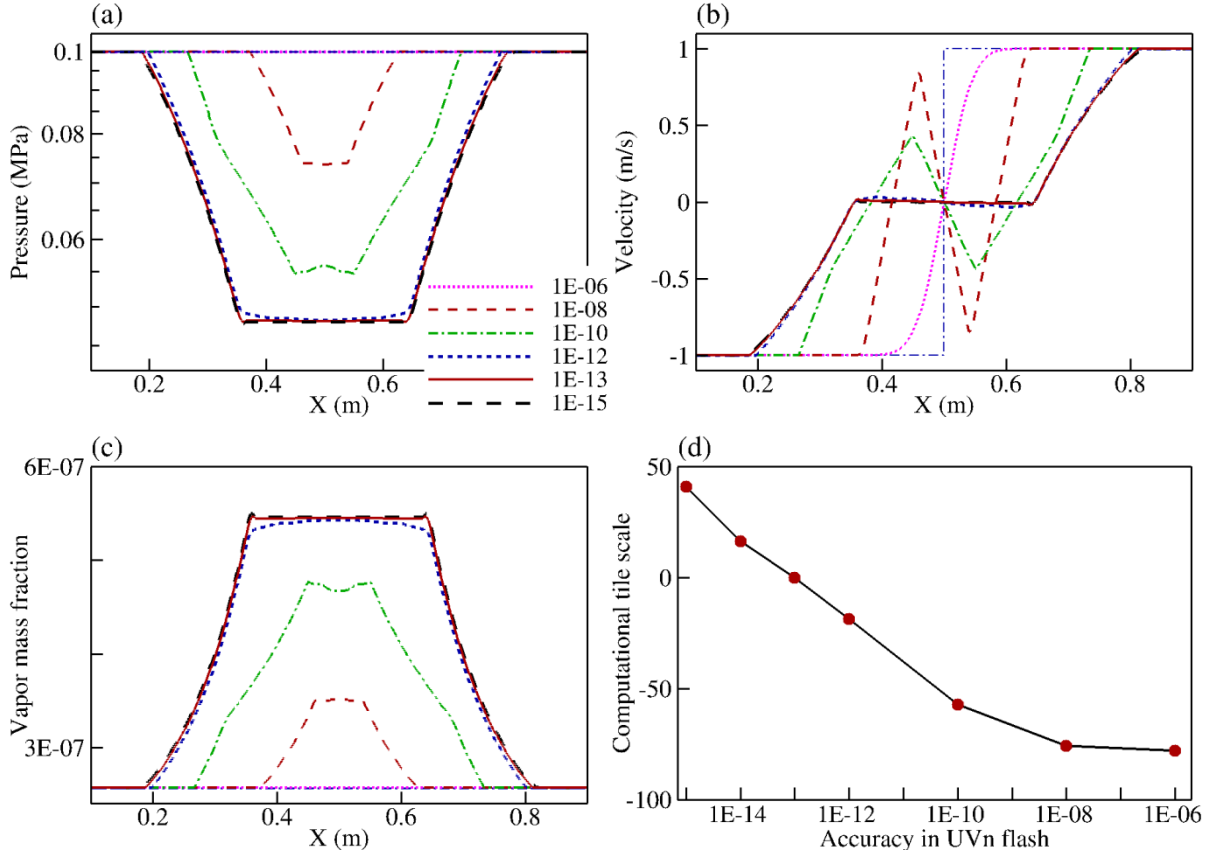


Fig. 10 Convergence analysis of the UVn flash solver (Case 2,  $t = 3.5$  ms), computational time scale =  $100 \times (t_\varepsilon - t_{\varepsilon=13})/t_{\varepsilon=13}$ , thin dash dot lines are initials, CFL = 0.15.

The double-expansion tube (Case 2 in Section 3.1.3) is simulated using the convergence tolerance ( $\varepsilon_{UVn}$ ) in the range of 1.0e-6 – 1.0e-15, and the results are presented in Fig. 10. It is found that there is no phase change during the expansion process with a large convergence tolerance of 1.0e-6. Both the expansion width and phase change quantity rise as the convergence tolerance decreases to the value of 1.0e-13. Moreover, substantial oscillations can be observed in the velocity profile with large convergence tolerance. Notably, the wave of pressure, velocity, as well as the phase change quantity are identical with the strict convergence tolerance of 1.0e-13 – 1.0e-15. Besides, the stricter convergence tolerance leads to longer computational time as expected and shown in Fig. 10(d).

1 Considering the compromise between the computational accuracy and efficiency, a convergence  
 2 tolerance of  $1.0e-13$  is chosen to get acceptable predictions in the cavitation tube.

#### 4. Discussion

5 In order to gain better understanding of the complex wave behaviors of *n*-dodecane/nitrogen mixture,  
 6 especially for the phase change between multicomponent liquid and gas phases, additional five cases  
 7 are computed, and the details are listed in Table 3 and Table 4.

##### 4.1 Behaviors of *n*-dodecane/nitrogen mixture in shock tubes

10 In this sub-section, the non-ideal behaviors of *n*-dodecane/nitrogen mixture in shock tubes are  
 11 discussed based on the numerical results. The tube is filled with high-pressure liquid in the left side  
 12 and low-pressure gas in the right side. The initial temperature throughout the tube is 293 K on both  
 13 sides. Two shock tube cases are simulated, one considers tiny discontinuities at the interface between  
 14 two sides (Case 4), and another case involves large discontinuities in feed (Case 5), as listed in Table 3.

16 Table 3. Initial conditions for shock tube cases with *n*-dodecane/nitrogen mixture

	$P_L$ (MPa)	$P_R$ (MPa)	$T_L = T_R$ (K)	$u_L = u_R$ (m/s)	$\alpha_{g,L}$ Equilib.	$\alpha_{g,R}$ Equilib.	$Y_{C_{12},L}$ Input	$Y_{C_{12},R}$ Input
Case 4	0.2	0.1	293	0	0.0117	0.1354	0.9995	0.9995
Case 5	10	0.1	293	0	0.0001	0.9999	0.9788013	0.0211987

##### 4.1.1 Results of Case 4

19 In Case 4, a mixture with a large amount of *n*-dodecane ( $Y_{C_{12}H_{26}} = 0.9995$ ) is selected as the feed.  
 20 Since the pressures in left and right sides are different, the initial gas volume fraction at equilibrium  
 21 state predicted by the TPn flash in left and right sides equals 0.0117 and 0.1354, respectively, which is  
 22 far from the phase boundaries. Fig. 11 shows the wave evolution behaviors of *n*-dodecane/nitrogen  
 23 mixture in the shock tube at  $t = 2.0$  ms. The conventional expansion and compression waves are  
 24 shown in Fig. 11(a), and the velocity magnitude depicted in Fig. 11(c) is low due to the high mixture  
 25 density and tiny gradients in thermodynamic properties. In addition, the large amount of liquid in Case  
 26 4 shows higher specific heat capacity, thereby the fluid is close to quasi-isothermal even though there  
 27 is a slight evaporation and condensation. This shock tube case highlights the accuracy of this four-  
 28 equation model when dealing with tiny gradients in thermodynamic properties.

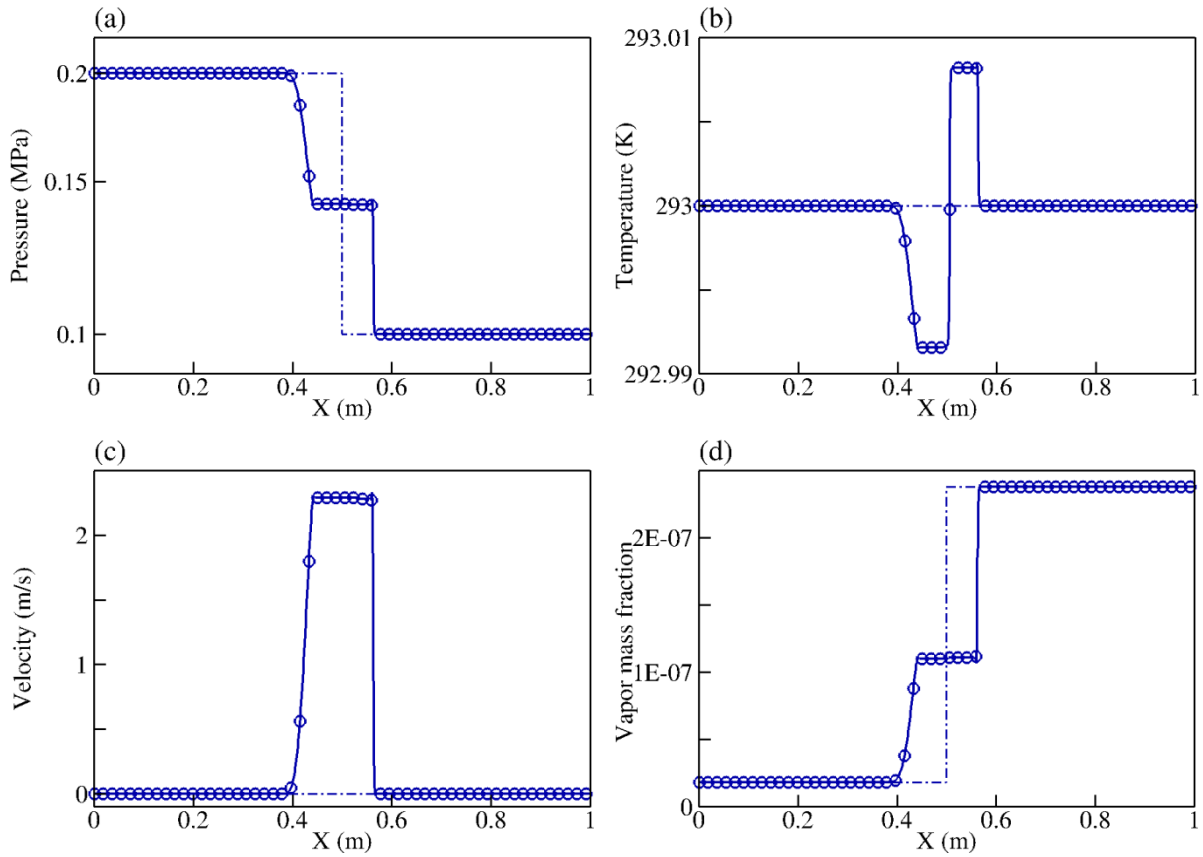


Fig. 11 Wave behaviors of *n*-dodecane/nitrogen mixture in the shock tube predicted by the four-equation model (Case 4:  $P_L = 0.2$  MPa,  $P_R = 0.1$  MPa,  $T_L = T_R = 293$  K,  $\alpha_{g,L} = 0.0117$ ,  $\alpha_{g,R} = 0.1354$ ,  $t = 2.0$  ms, thin dash dot lines are initials).

#### 4.1.2 Results of Case 5

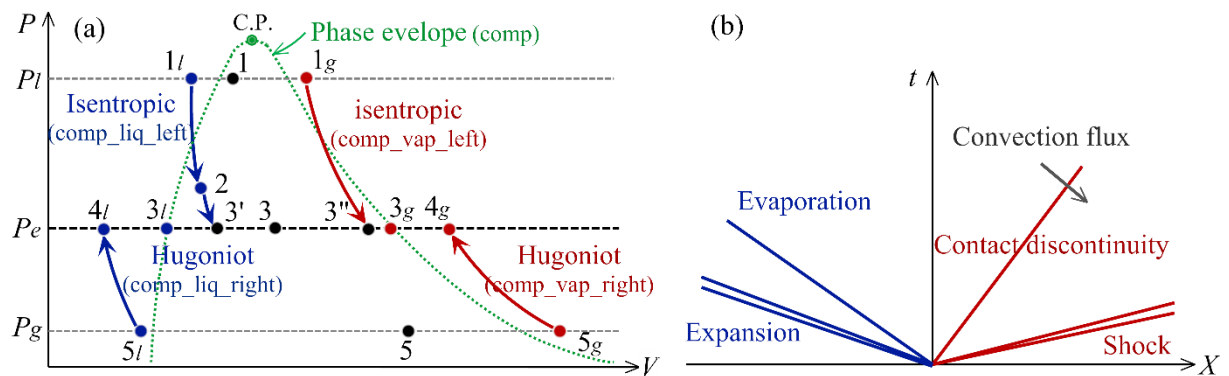


Fig. 12 Phase trajectories and wave patterns for fluids (a) phase trajectories; (b) wave patterns ( $z$ : mixture composition;  $y_{k\_left}$ : gas composition at left;  $y_{k\_right}$ : gas composition at right;  $x_{k\_left}$ : liquid composition at left;  $x_{k\_right}$ : liquid composition at right).

1 In this case, the input mass fraction of *n*-dodecane in left and right sides of the shock tube diaphragm  
2 is 0.9788013 and 0.0211987, respectively. Through the TPn flash, the initial equilibrium  $\alpha_g$  in left and  
3 right sides is determined as 0.0001 and 0.9999, respectively. The wave evolution profiles of this  
4 mixture at the instant of 1.0 ms are plotted in Fig. 13. As the discontinuities of feed at the interface are  
5 increased relatively to those in Case 4, the wave behaviors of *n*-dodecane/nitrogen mixture in the  
6 shock tube shown in Fig. 13 are more complex than those in Case 4 (Fig. 11). It can be seen that the  
7 current model can capture the essential features of the mixture with large gradients in thermodynamic  
8 properties. Specifically, four waves can be observed: the left expansion wave, evaporation front,  
9 contact discontinuity, and the right shock wave. The detailed wave patterns and phase trajectories are  
10 presented in Fig. 12.

11  
12 Following the analysis of (Saurel et al., 2008), the mixture Hugoniot curve is tangent to their isentrope,  
13 and the multiphase shock waves behave like simple compression waves. Therefore, the phase  
14 trajectory of this case with large discontinuities of density can follow the phase schematic of Fig. 12(a).  
15 In the left high-pressure side, the initial equilibrium mixture represented by point 1 is composed of  
16 large amount of liquid (point  $1_l$ ) and a bit gas (point  $1_g$ ). As time begins, an expansion wave is  
17 generated and travels to the left side with the velocity of  $u - C_{s,mix}$ , the pressure falls rapidly across  
18 the expansion wave. The mass fraction of dissolved nitrogen follows similar evolution behaviors with  
19 the pressure as shown in Fig. 13(a) and (b). As known that the dissolved nitrogen is a kind of dense-  
20 phase fluid with high vapor pressure, which dramatically enhances the phase change and expansion  
21 intensity (Kuijpers et al., 2002). Therefore, it cannot be neglected in the simulation. The initial liquid  
22 indicated by point  $1_l$  is expanded to the metastable liquid (point 2) following an isentropic path. In the  
23 present modelling, this metastable liquid is relaxed to a thermodynamic equilibrium mixture (point  $3'$ )  
24 through the UVn flash. Thus, the evaporation front appears and travels together with the expansion  
25 wave to the left high-pressure side. Both *n*-dodecane vapor and nitrogen gas are produced as shown in  
26 Fig. 13(e). The initial gas of point  $1_g$  is expanded following an isentropic path to the point  $3''$ , which is  
27 relaxed to the thermodynamic equilibrium mixture (points  $3_g$  and  $3_l$ ) infinitely. The corresponding  
28 densities of the mixtures represented by points 1 and 3 ( $3 = 3' + 3''$ ) are reported in Fig. 13(f).

29

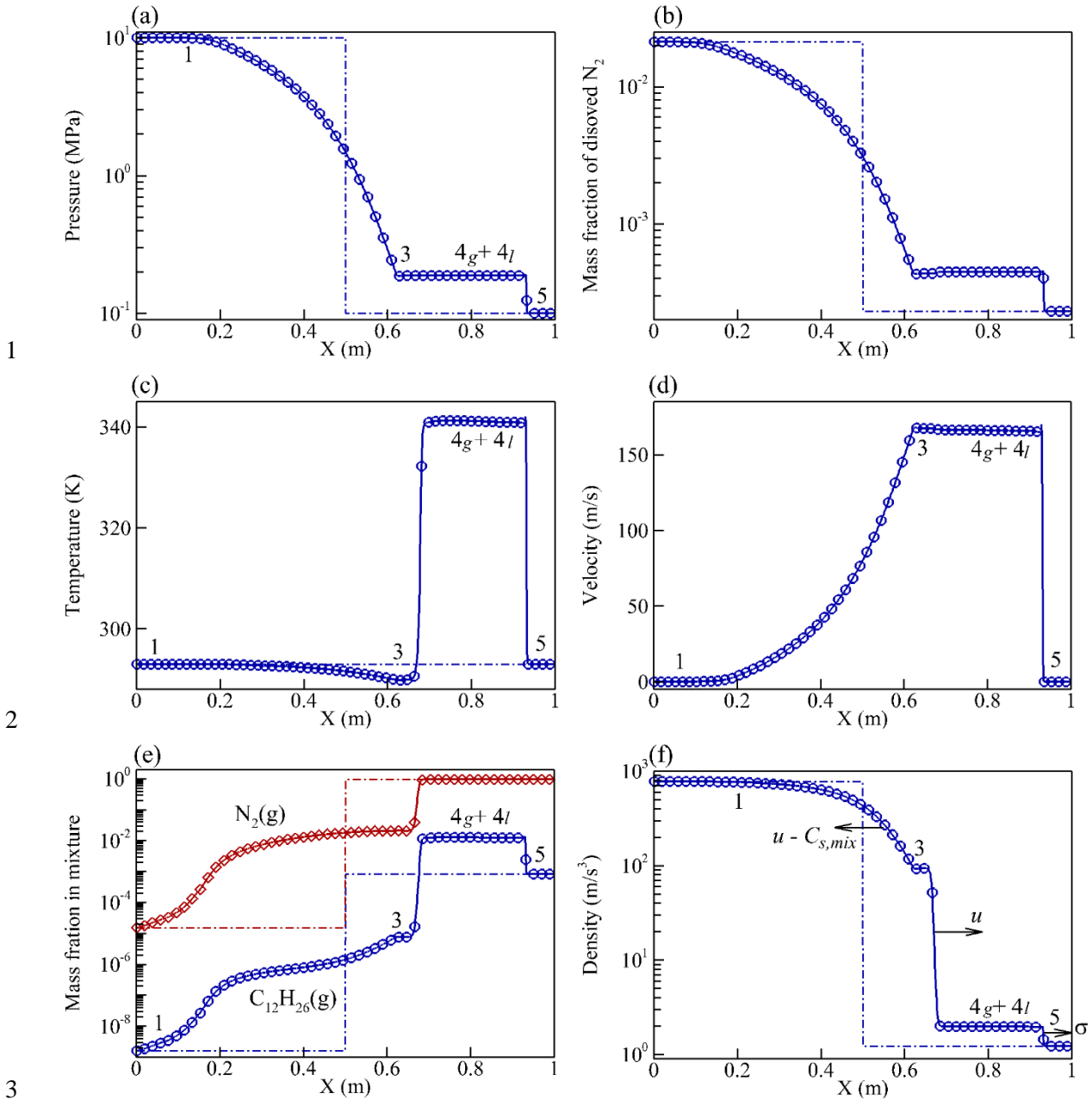


Fig. 13 Wave behaviors of *n*-dodecane/nitrogen mixture in the shock tube predicted by the four-equation model (Case 5:  $P_L = 10.0$  MPa,  $P_R = 0.1$  MPa,  $T_L = T_R = 293$  K,  $\alpha_{g,L} = 0.0001$ ,  $\alpha_{g,R} = 0.9999$ ,  $t = 1.0$  ms, thin dash dot lines are initials)

Since there are large discontinuities in densities and species at the contact interface, the liquid at interface undergoes a sudden large phase change in a narrow region (Simões-Moreira, 2000), and the vapor is ejected from the interface and propagated to the right low-pressure side through the convection flux at the velocity of  $u$  (Fig. 12(b) and Fig. 13(f)), resulting in an increase in vapor mass fraction (Fig. 13(e)). In the right low-pressure side, the equilibrium mixture of point 5 is composed of large amount of gas ( $5_g$ ) and small amount of liquid ( $5_l$ ). At beginning, the shock wave is yielded and propagates to the right low-pressure side at the velocity of  $\sigma$ . The gaseous mixture ( $5_g$ ) is shocked following a

1 Hugoniot curve (Saurel et al., 2008) from point  $5_g$  to  $4_g$  as shown in Fig. 12(a). Thus, the pressure,  
 2 temperature, velocity and the mixture density are increased. Since points 3 and  $4_g$ , as well as  $4_l$  have  
 3 no thermodynamic connections (Fig. 12(a)), they are only linked by the mechanical equilibrium  
 4 through a simple contact discontinuity separating the evaporated vapor front and shocked mixtures.  
 5 Thereby, the pressure and velocity are continuous, while, a sharp contact discontinuity is generated in  
 6 the profiles of temperature, vapor mass fraction, and density between the mixtures of points 3 and  
 7  $4_g + 4_l$ . The slopes of left expansion and right shock waves can be expressed as  $-\rho C_{s,mix}$  and  
 8  $\rho(\sigma - u)$ , respectively. It is noted that the left slop is much lower than the right one (Fig. 13(c)). This  
 9 is because the sound speed in two-phase mixtures ( $C_{s,mix}$ ) decreases progressively along the expansion  
 10 evaporation front, following the Wood formula (Wood, 1930). Finally, since the mixture is proven to  
 11 be in two-phase state during the whole process, no wave splitting has been obtained in the expansion  
 12 wave.

13

#### 14 4.2 Behaviors of *n*-dodecane/nitrogen mixture in double-expansion tubes

15 In this subsection, three double-expansion tube cases are simulated, and the details are listed in Table  
 16 4. In Section 4.2.1, the influence of volume translation in PR-EoS on liquid density is presented. Then,  
 17 its influence on the expansion behaviors of *n*-dodecane/nitrogen mixture is discussed in the following  
 18 Sections 4.2.2 – 4.2.4, in which the expansion characteristics under various conditions are included.

19

20 Table 4. Initial conditions for double-expansion tube cases with *n*-dodecane/nitrogen mixture

	$p_L = p_R$ (MPa)	$T_L = T_R$ (K)	$u_L$ (m/s)	$u_R$ (m/s)	$\alpha_{g,L} = \alpha_{g,R}$ Equilib.	$Y_{c12,L} = Y_{c12,R}$ Input
Case 6	0.1	293	-1	1	1.0e-04	0.9997697
Case 7	0.1	480	-1	1	1.0e-04	0.9999349
Case 8	5.0	293	-1	1	1.0e-04	0.9889499

21

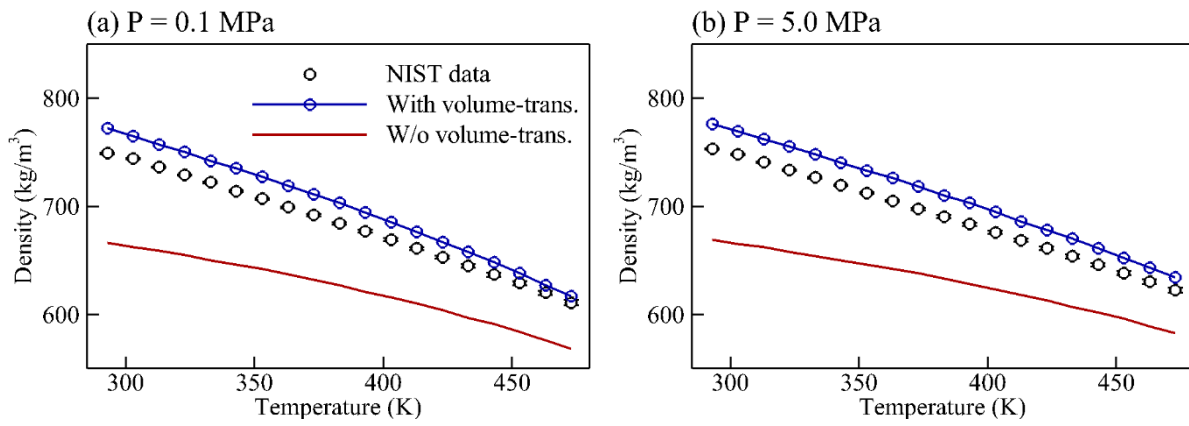
##### 22 4.2.1 Influence of volume translation in PR-EoS on density and sound speed

23 The liquid density makes substantial influence on the prediction accuracy for expansion behaviors of  
 24 the *n*-dodecane/nitrogen mixtures. Since the liquid density of hydrocarbons computed by the PR EoS  
 25 is known to be inaccurate, the volume translation (Baled et al., 2012, Tapriyal et al., 2012) is added to  
 26 the PR EoS to improve the prediction accuracy of liquid density as explained in Section 2.1. Results  
 27 with and without the volume translation for liquid *n*-dodecane density under various temperature and  
 28 pressure conditions are compared in Fig. 14(a). The test temperature and pressure are in the range of  
 29 293 – 480 K and 0.1 – 5.0 MPa, respectively. It can be seen that the model without volume translation  
 30 substantially underestimates the liquid density compared to NIST data (Linstrom and Mallard, 2001).  
 31 Comparatively, the predictions with volume translation are improved significantly.

32

1 Since the dissolved nitrogen quantity in liquid phase is increased with pressure, the effects of  
 2 dissolved nitrogen on the liquid thermal properties under different pressure conditions need to be  
 3 illustrated. The compared density and sound speed between the predictions and NIST data at pure  
 4 liquid state ( $TPD = 1$  and  $\alpha_g = 1.0e-06$ ) are illustrated in Fig. 15. The mass fraction of dissolved  
 5 nitrogen in liquid phase varies from  $1.0e-6$  to  $1.0e-4$ . It is found that the liquid density and sound  
 6 speed are insensitive to the dissolved nitrogen quantity at 0.1 MPa condition. However, at the high  
 7 pressure of 5.0 MPa, both of liquid density and sound speed decrease as the dissolved nitrogen  
 8 increases. Thereby, it can be concluded that the dissolved nitrogen quantity in the liquid phase is  
 9 important for the simulation accuracy at high pressure conditions.

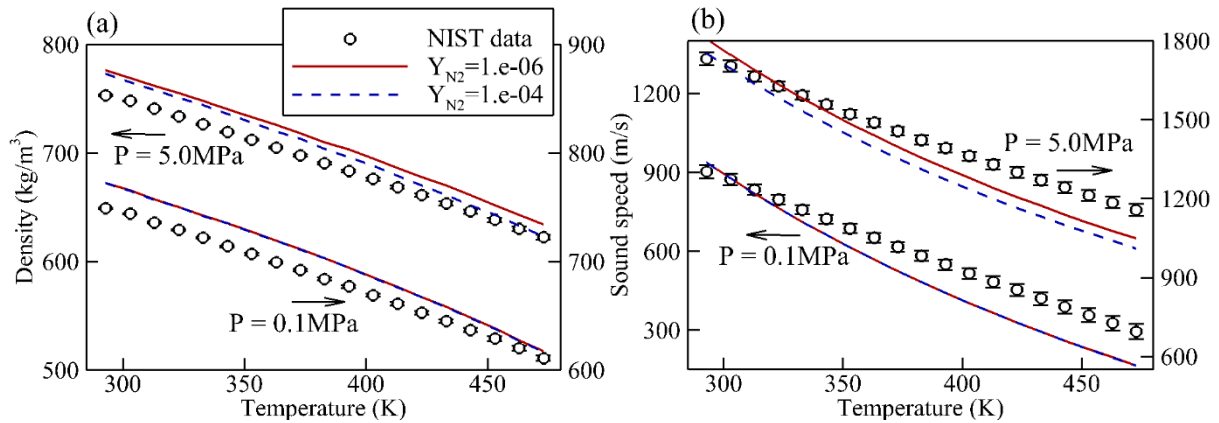
10



11

12 Fig. 14 Liquid density of *n*-dodecane predicted by PR-EoS with and without volume translation (circle:  
 13 data from NIST (Linstrom and Mallard, 2001),  $\alpha_g = 1.0e-06$ ,  $Y_{N_2} = 1.0e-04$ )

14



15

16 Fig. 15 Effects of the dissolved nitrogen in liquid phase on density and sound speed (circle: data from  
 17 NIST (Linstrom and Mallard, 2001) for pure liquid *n*-dodecane,  $\alpha_g = 1.0e-06$ )

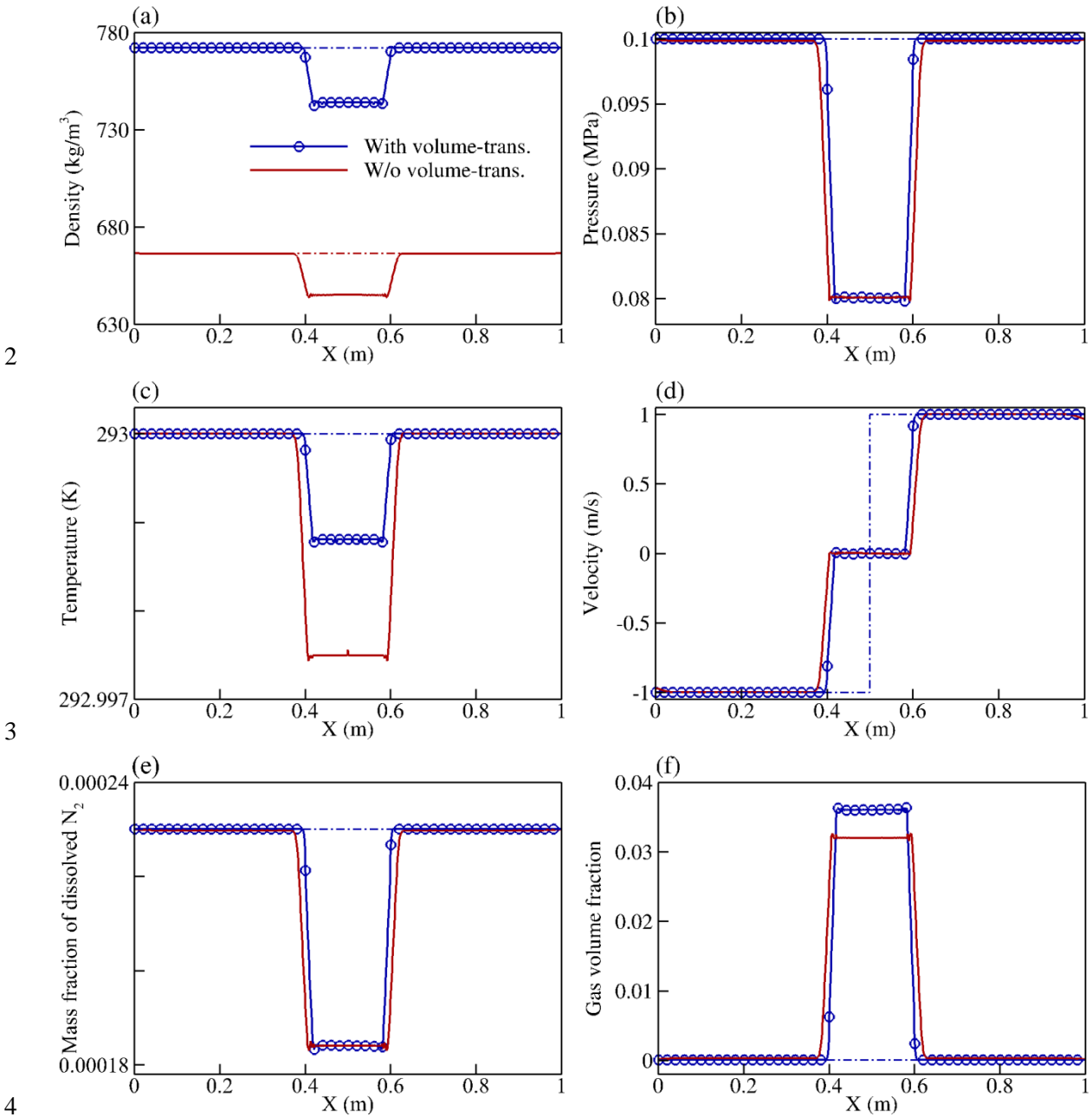
18

19

20

21

1 4.2.2 Results of Case 6



5 Fig. 16 Wave behaviors of *n*-dodecane/nitrogen mixture in the double-expansion tube predicted by the  
 6 four-equation model with and without volume translation (Case 6:  $P_L = P_R = 0.1$  MPa,  $T_L = T_R =$   
 7 293 K,  $\alpha_{g,L} = \alpha_{g,R} = 0.0001$ ,  $u_L = -1.0$  m/s,  $u_R = 1.0$  m/s,  $t = 3.5$  ms, thin dash dot lines are  
 8 initials)

9  
 10 The wave patterns of density, speed of sound, pressure, temperature, dissolved nitrogen, as well as the  
 11 gas phase volume fraction in the double-expansion tube with initial pressure and temperature of 0.1  
 12 MPa and 293 K are presented in Fig. 16. The initial velocity of fluid in the left and right sides are -1  
 13 and +1 m/s, respectively. As expected, a large discrepancy is shown in the initial density. Ignoring the  
 14 volume translation underestimates the liquid density about 13%, resulting in a higher expansion rate,



1 and the expanded strength is slightly larger than that predicted by the model with volume translation.  
2 A little cooling effect caused by the phase change is captured in the temperature profile. Discrepancies  
3 are observed in the evolution of temperature and gas volume fraction predicted by the models with and  
4 without the volume translation as shown in Fig. 16(d) and (f). This is due to the heat balance between  
5 two phases, the temperature drop and gas volume fraction are estimated by  $\Delta T = (\rho_g L_{evap}) / (\rho_l C_{p,l})$   
6 and  $\alpha_g = 1 / [(\rho_g L_{evap} / \Delta T \rho_l C_{p,l}) + 1]$  (Michel and Franc, 2004). It is found that the liquid density  
7 predicted by the model with volume translation is higher than that without volume translation,  
8 resulting in a decrease in the temperature drop and an increase in the gas volume fraction.

#### 10 4.2.3 Results of Case 7

11 The primary aim of Case 7 is to investigate the influence of temperature on the expansion behaviors  
12 for the *n*-dodecane/nitrogen mixture. Fig. 17 presents the profiles of density, pressure, temperature,  
13 velocity, dissolved nitrogen, and gas phase volume fraction at 3.5 ms, the initial pressure and  
14 temperature of the fluid in the double-expansion tube are 0.1 MPa and 480 K, which is quite close to  
15 the boiling temperature of pure *n*-dodecane. Compared to the results of Case 6 with low initial  
16 temperature (293 K in Fig. 16), several conclusions can be obtained. First, the liquid density decreases  
17 with the temperature increasing. Second, the expanded width is substantially reduced, whereas the  
18 cooling effect by the phase change is enhanced by the high temperature. Third, the solubility of  
19 nitrogen in liquid phase decreases with an increase in temperature as expected. The mass fraction of  
20 nitrogen dissolved in the liquid phase in Case 7 varies from 3.0e-05 to 6.5e-05 (Fig. 17(e)), which is  
21 10 times lower than that in Case 6 with low initial temperature (Fig. 16(e)). In addition, since the  
22 liquid vapor pressure increases with temperature, the gas content in the cavitation region is increased  
23 as shown in Fig. 17(f).

25 Based on the results of Fig. 17 for Case 7, the density deviation predicted by the model without  
26 volume translation is around 7.5%, which is less than that under the conditions with low initial  
27 temperature (around 13% in Case 6). Besides, the evolution of pressure, temperature and velocity  
28 predicted by the models with and without volume translation are quite close. Thereby, it can be  
29 concluded that the volume translation method shows insignificant effects on the expansion behaviors  
30 in Case 7. In other words, it demonstrates that the expansion characteristics of *n*-dodecane/nitrogen  
31 mixture is non-sensitive to the liquid density at high temperature conditions.

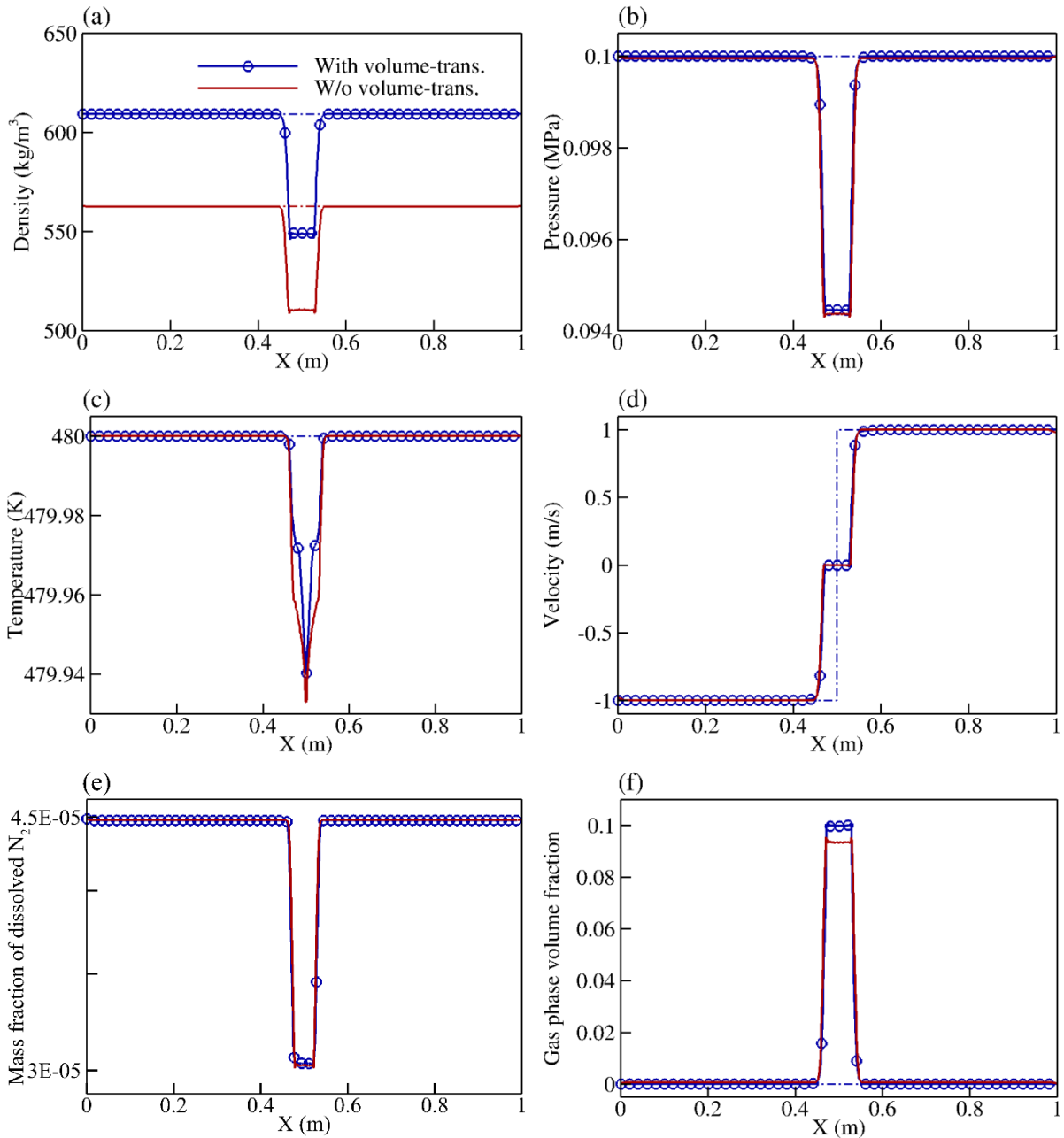
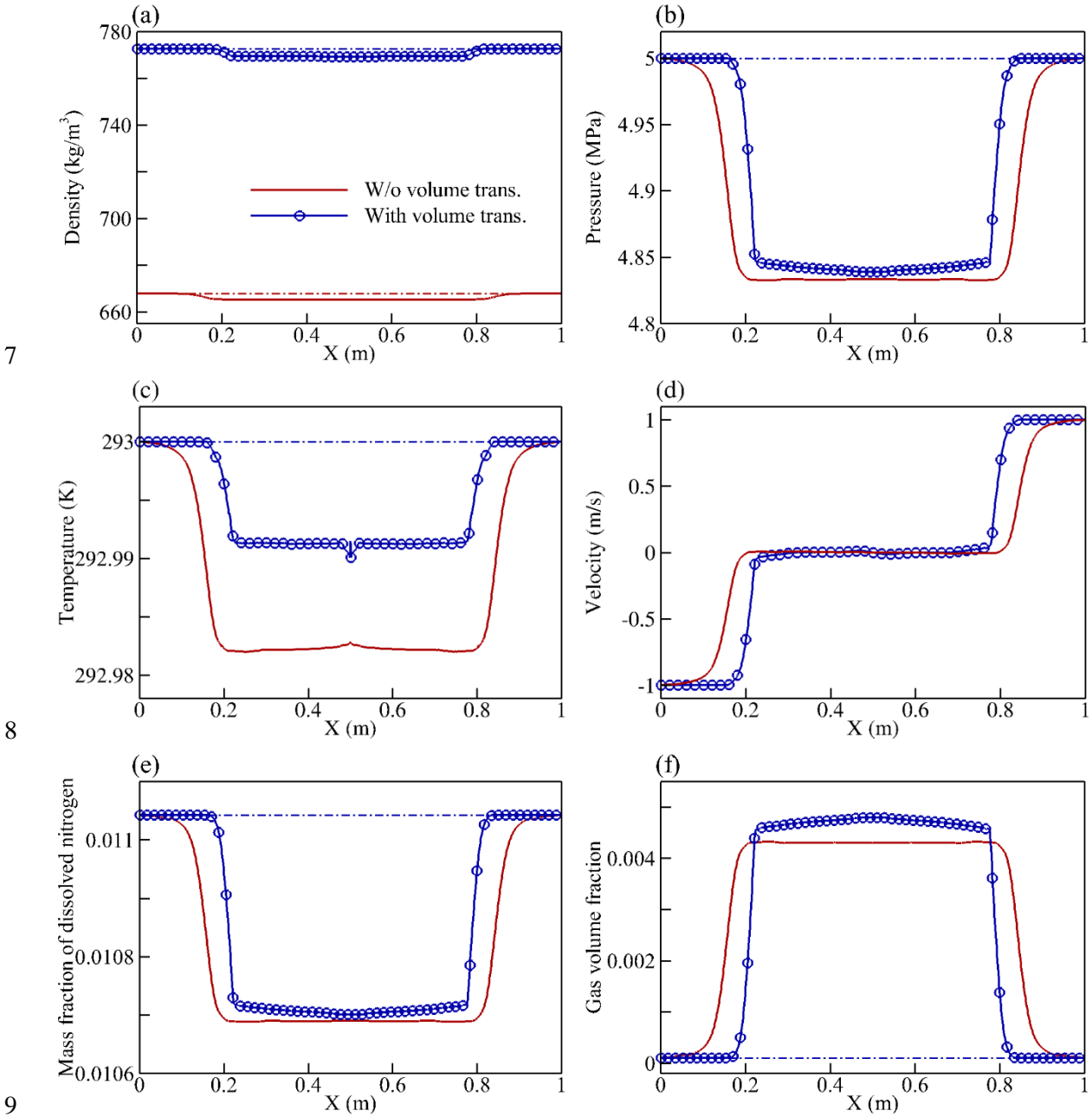


Fig. 17 Wave behaviors of *n*-dodecane/nitrogen mixture in the double-expansion tube predicted by the four-equation model with and without volume translation (Case 7:  $P_L = P_R = 0.1$  MPa,  $T_L = T_R = 480$  K,  $\alpha_{g,L} = \alpha_{g,R} = 0.0001$ ,  $u_L = -1.0$  m/s,  $u_R = 1.0$  m/s,  $t = 3.5$  ms, thin dash dot lines are initials)

#### 4.2.4 Results of Case 8

Another important variable for expansion behaviors is pressure. The wave evolution profiles of *n*-dodecane/nitrogen mixture in the double-expansion tube are presented in Fig. 18. The initial temperature and pressure of fluid are 293 K and 5.0 MPa. Compared to the results of Case 6 with low initial pressure (0.1 MPa), the expanded speed and the dissolved nitrogen quantity are increased remarkably in Case 8 with high initial pressure (5.0 MPa). The mass fraction of dissolved nitrogen in

1 the liquid phase is seen to be as large as 0.01105 (see Fig. 18(e)), since the dissolved nitrogen possess  
 2 a high vapor pressure, it can improve the phase change and expansion intensity a lot (Kuijpers et al.,  
 3 2002). Thereby, it can be concluded that the multicomponent two-phase flow model, such as this four-  
 4 equation model, is strongly required for accurately predicting the homogeneous nucleation during the  
 5 pressure expansion processes, like in diesel and GDI engines with high injection pressures.  
 6



7  
 8  
 9  
 10 Fig. 18 Wave behaviors of *n*-dodecane/nitrogen mixture in the double-expansion tube predicted by the  
 11 four-equation model with and without volume translation (Case 8:  $P_L = P_R = 5.0$  MPa,  $T_L = T_R =$   
 12  $293$  K,  $\alpha_{g,L} = \alpha_{g,R} = 0.0001$ ,  $u_L = -1.0$  m/s,  $u_R = 1.0$  m/s,  $t = 1.4$  ms, thin dash dot lines are  
 13 initials).

1 As the pressure increases, the volume translation added in PR EoS becomes very important in density  
 2 predictions and subsequently influences the expansion behaviors. The compared results between the  
 3 models with and without volume translation are plotted in Fig. 18. Large deviation can be observed in  
 4 the expansion width predicted by the model without volume translation. Therefore, the volume  
 5 translation is demonstrated to be important in the real-fluid simulation using PR EoS, especially at  
 6 high pressure conditions.

### 7 8 5. Three-dimensional n-dodecane injection modelling

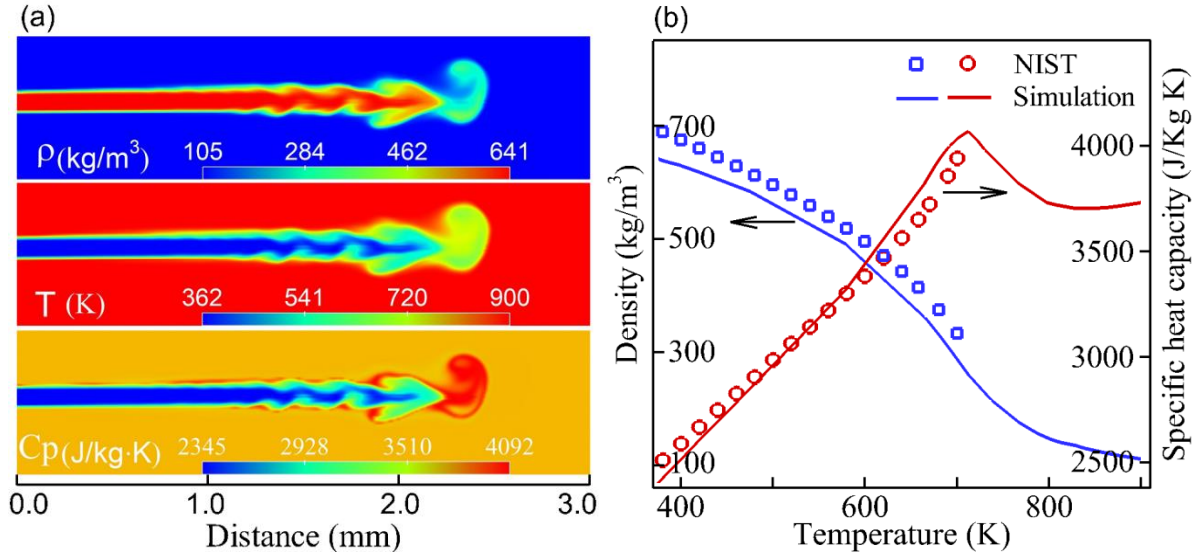
9 To study the performance of the proposed four-equation model for multidimensional flows, a 3D  
 10 transcritical fuel injection is simulated. Here, the transcritical injection is defined as that both injection  
 11 and chamber pressures exceed the fluid critical pressure, and the injection temperature is lower but the  
 12 chamber temperature is higher than the fluid critical temperature (Ma et al., 2017). Therefore, the  
 13 injected fluid follows a path crossing the pseudo-boiling line and evolves from a liquid-like state to a  
 14 gas-like state at  $T_{pseudo}$  as reported recently by (Banuti, 2015) and (Yang et al., 2018). In this  
 15 simulation, the phase change is not expected. The computational domain is illustrated in Fig. 19(a). A  
 16 typical injector which consists of a single-hole (Length = 1 mm and Diameter = 100  $\mu\text{m}$ ) is fitted to a  
 17 hexahedral chamber. The boundary conditions are set with pressure inlet and outlet in left and right  
 18 sides of the geometry, respectively, as shown in Fig. 19(b). A liquid *n*-dodecane (index  $C_{12}$ ) jet at 363  
 19 K ( $T_{jet} < T_{c,C_{12}} = 658.1$  K) is injected into a chamber filled with vapor *n*-dodecane at the temperature  
 20 of 900 K ( $T_{chamber} > T_{c,C_{12}}$ ). The injector and chamber pressures are initially set to be 7 and 4 MPa,  
 21 respectively, which are above the *n*-dodecane critical pressure ( $P_{c,C_{12}} = 1.82$  MPa).



23  
24 Fig. 19 Schematic and mesh distribution of a typical injector (a) Schematic; (b) Mesh (204800 cells,  
25 the minimum size is 10  $\mu\text{m}$ ).

26  
27 The density, temperature, and isobaric heat capacity behaviors at the instant of 0.09 ms are plotted in  
 28 Fig. 20(a). The jet is evolving from a liquid-like supercritical state ( $T < T_{pseudo}$ ) to a gas-like  
 29 supercritical state ( $T > T_{pseudo}$ ). The fluid thermodynamic state is correctly predicted as shown in Fig.  
 30 20(b). Indeed, the predicted density and isobaric heat capacity agree very well with the available NIST  
 31 data (Linstrom and Mallard, 2001). In particular, the non-linearity of isobaric across the pseudo-  
 32 boiling line seems to be well captured by the PR EoS. This phenomenon appears at the jet periphery  
 33 and also in the jet front (see  $C_p$  in Fig. 20(a). The peak value of isobaric heat capacity is very large  
 34 (around 4  $\text{kJ}/(\text{kg} \cdot \text{K})$ ), which may have significant cooling effects on the temperature distribution

1 before combustion. Of course, this kind of non-linearity behavior cannot be obtained by the ideal gas  
 2 EoS (Segal and Polikhov, 2008). Therefore, to better quantify the performance of transcritical  
 3 injection and combustion, the real-fluid two-phase flow model should be considered.



5  
 6 Fig. 20 Results of injection event (a) temporal sequence of density ( $\rho$ ), temperature ( $T$ ), and isobaric  
 7 heat capacity ( $C_p$ ); (b) comparison between predicted density and isobaric heat capacity with available  
 8 NIST data (Here, the density and heat capacity varied with temperature are in the radial section at the  
 9 distance of 1.85 mm from the hole exit), ( $P_{jet} = 7.0$  MPa,  $T_{jet} = 363$  K,  $P_{chamber} = 4.0$  MPa,  
 10  $T_{chamber} = 900$  K,  $Y_{C_{12}H_{26}} = 0.99999$ ,  $t = 0.09$  ms).

## 12 6. Conclusions

13 A multicomponent fully compressible four-equation model with a real-fluid equilibrium-solver was  
 14 constructed and implemented into an in-house IFP-C3D software. It was validated against the  
 15 experimental results, and the capability to deal with two-phase flows was highlighted in both 1D and  
 16 3D test cases. According to the numerical results, the following conclusions can be obtained.

- 18 (1) The suggested four-equation model can compute the real-fluid phase change for multicomponent  
 19 two-phase flows, and it can also predict more accurate real-fluid behaviors, including the effects of  
 20 dissolved nitrogen in liquid phase;
- 21 (2) The “composite EoS” approach using respective PR EoS in their range of convexity for liquid and  
 22 gas phases, and connected in the vapor dome by a set of equilibrium constraints, can solve the  
 23 drawback of “mixture PR EoS” in the unstable spinodal region, and so preserve the hyperbolicity  
 24 of the Euler system;

- 1 (3) Referring to the wave behaviors of water-nitrogen in the 1D shock and double-expansion tubes  
 2 with tiny discontinuities at the interface, the predictions of present four-equation model are close  
 3 to those of Chiapolino et al. (2017) model;
- 4 (4) The flash boiling process of *n*-dodecane/nitrogen mixture is satisfactorily reproduced by the  
 5 present four-equation model, and the predicted results show good agreements with experimental  
 6 measurements. The complex wave patterns in flash boiling cases are successfully tracked;
- 7 (5) Based on the sensitivity analysis, this four-equation model can illustrate reliable and efficient  
 8 calculations with a convergence criterion of 1.0e-13 and a CFL number in the range of 0.15 – 0.3;
- 9 (6) This four-equation model can produce the complex wave behaviors satisfactorily, including the  
 10 expansion, evaporation and shock fronts, as well as the contact discontinuity regime for *n*-  
 11 dodecane/nitrogen mixtures with large discontinuities at the interface. The wave evolutions have  
 12 been revealed and analyzed based on the thermodynamic phase trajectories under various  
 13 conditions;
- 14 (7) The dissolved nitrogen quantity in liquid phase becomes larger as the pressure increases and  
 15 temperature decreases, its evolution profile is quite close to the pressure. The phase change and  
 16 cavitation intensity are improved with large amount of dissolved nitrogen in the liquid phase;
- 17 (8) The volume translation in PR EoS shows negligible effects on the liquid density, expansion  
 18 behaviors for the *n*-dodecane/nitrogen mixture in the cases with high initial temperature. However,  
 19 as the initial pressure increases and temperature decreases, the volume translation model may help  
 20 to obtain the correct density and expansion behaviors.
- 21 (9) Finally, the suggested four-equation model has been shown to be able of predicting the main  
 22 features of a typical 3D subcritical and transcritical injections.

23

24 Declarations of interest: none

25

26 Acknowledgement

27 This project has received funding from the European Union Horizon 2020 Research and Innovation  
 28 programme. Grant Agreement No 675528.

29

30 Appendix A. Thermodynamic properties and equilibrium-solver

31 A.1. Thermodynamic properties of single-phase

32 Thermodynamic parameters in this study are evaluated consistently with PR EoS, including the  
 33 pressure, specific internal energy, density, and the fugacity of species:

$$34 \quad \left. \frac{\partial P}{\partial T} \right|_{\rho} = \frac{R}{V-b} - \frac{da}{dT} \frac{1}{V^2+2bV-b^2} \quad (\text{A. 1})$$

$$35 \quad \left. \frac{\partial P}{\partial \rho} \right|_T = -\frac{M}{\rho^2} \left[ \frac{-RT}{(V-b)^2} + \frac{2a(V+b)}{(V^2+2bV-b^2)^2} \right] \quad (\text{A. 2})$$

$$e_d = \left. \frac{\partial e}{\partial T} \right|_P = \left( T \frac{da}{dT} - a \right) \ln \left( \frac{V + (1 + \sqrt{2})b}{V + (1 - \sqrt{2})b} \right) / (2\sqrt{2}bM) \quad (\text{A. 3})$$

$$e = e_0 + e_d \quad (\text{A. 4})$$

$$\left. \frac{\partial \rho}{\partial T} \right|_P = - \frac{\partial P}{\partial T} \left( \left. \frac{\partial P}{\partial \rho} \right|_T \right)^{-1} \quad (\text{A. 5})$$

$$f_k = x_k \phi_k \quad (\text{A. 6})$$

$$\phi_k = \exp \left( \left( \frac{b_k}{B} \right) (Z - 1) - \ln(Z - B) + \left( \frac{A}{2\sqrt{2}B} \right) \left[ \frac{b_k}{B} - \frac{2.0}{A\alpha} \sum_{k'=1}^N x_{k'} a_{k'} \alpha_{kk'} \right] \ln \left( \frac{Z + 2.414B}{Z - 0.414B} \right) \right) \quad (\text{A. 7})$$

where  $e$  is the internal energy, which is computed from the sum of the departure part of  $e_d$  (Vidal, 2003) and the ideal gas part  $e_0$ , which is calculated based on the polynomial equation (Aly and Lee, 1981).  $\phi_k$  is fugacity coefficient;  $Z$  is compressibility factor, calculated as

$$Z^3 - (1 - B)Z^2 + (A - 3B^2 - 2B)Z - (AB - B^2 - B^3) = 0 \quad (\text{A. 8})$$

where  $A = aP/R^2T^2$  and  $B = bP/RT$ ; the coefficients  $a$  and  $b$  are estimated based on the van der Waals mixing rule as  $a = \sum \sum x_{k_1} x_{k_2} a_{k_1 k_2}$ ,  $b = \sum x_{k_1} b_{k_1}$ ,  $a_k$  and  $b_k$  are determined based on the critical point of each component (Kwak and Mansoori, 1986),  $k_{k_1 k_2} = 0.19$  for  $n$ -dodecane/nitrogen and  $k_{k_1 k_2} = 0.0$  for water-nitrogen in this study.

## A.2. Phase stability test

The phase stability test is an important part of phase equilibrium computation. It is used to decide whether the system is thermodynamically stable. In this study, the Tangent Plane Distance (TPD) criterion (Michelsen, 1982) is used. In this approach, the TPD represents the vertical distance from the tangent hyperplane of Gibbs free energy surface at the feed  $z_k$  to that at the phase compositions  $x_k$ . The system stability requires the TPD function to be non-negative:

$$TPD = \sum_k^N x_k [\mu_k(x_k) - \mu_{k,0}(z_k)] \geq 0 \quad (\text{A. 9})$$

where  $x_k$  denotes the mole fraction of component  $k$  in one assumed phase. If non-negative TPD value is obtained with any trial phase compositions  $x_k$ , the initial mixture is stable and no phase split is needed. Otherwise, the initial mixture with the feed  $z_k$  is unstable, then the phase split happens. In this study, the label  $TPD = N_{phase}$  is used to identify the nature of phase with  $N_{phase} = 0$  for pure gas,  $N_{phase} = 1$  for pure liquid and  $N_{phase} = 2$  for two-phase.

## A.3. Isoenergetic-Isochoric (UVn) flash

Assuming the instantaneous thermodynamic equilibrium in each control volume, an isoenergetic-isochoric (UVn) flash is used to relax the Gibbs free energy of each phase. The mixture specific internal energy and density ( $e^*, \rho^*$ ) are known from the flow-solver (Eqs. (1) – (4)). The temperature, pressure, equilibrium factor, and vapor fraction ( $T, P, K_k, \psi_g$ ) are iterated based on the equilibrium solver (Eqs. (11) – (14)). The UVn flash iteration is composed of two parts: (1) Inner loop (TPn flash) and (2) Outer loop.

### A.3.1 Inner loop – Isothermal-isobaric (TPn) flash

1 In the TPn flash (Saha and Carrolls, 1997), the phase compositions ( $x_k, y_k$ ) are the target variables,  
 2 but they are not independent, the equilibrium factor ( $K_k$ ) and vapor fraction ( $\psi_g$ ) are. The equilibrium  
 3 factor ( $K_k$ ) is defined as

$$4 \quad K_k = y_k/x_k \quad (\text{A. 10})$$

5 where the phase compositions can be described as

$$6 \quad y_k = z_k K_k / [1 + (K_k - 1)\psi_g] \quad (\text{A. 11})$$

$$7 \quad x_k = z_k / [1 + (K_k - 1)\psi_g] \quad (\text{A. 12})$$

8 where  $\sum_{k=1}^N y_k = \sum_{k=1}^N x_k = 1$ . The objective functions in TPn flash are Rachford-Rice equation  
 9 (Saha and Carrolls, 1997) and fugacity equality equation described as

$$10 \quad F_1 = \sum_{k=1}^N z_k (K_k - 1) / [1 + \psi_g (K_k - 1)] \quad (\text{A. 13})$$

$$11 \quad F_2 = f_{k,l} / f_{k,g} - 1 \quad (\text{A. 14})$$

12 First, the objective function of Eq. (A. 13) is iteratively solved to obtain the vapor fraction  $\psi_g$ . Then,  
 13 the second objective function (A. 14) is solved to get the equilibrium constant ( $K_k$ ). The inner loop is  
 14 iterated until the convergence tolerance is fulfilled as Eq. (21). Otherwise, Eqs. (A. 13) and (A. 14) are  
 15 solved again with the new estimates for  $\psi_g$  and  $K_k$ . After the TPn flash, we can obtain the vapor mass  
 16 fraction and equilibrium constant ( $\psi_g$  and  $K_k$ ). The phase compositions ( $x_k, y_k$ ) can be determined  
 17 based on Eqs. (A. 11) – (A. 12).

18

19 The initializations of  $\psi_g$  and  $K_k$  are given by the solutions of flow-solver at current time-step  $n$ , as

$$20 \quad \psi_{g,n\_UVn^0} = \psi_{g,n}'' = m_{g,n}'' / (M_g \sum n_{k,g}'') \quad (\text{A. 15})$$

$$21 \quad K_{k,n\_UVn^0} = \phi_{k,l}'' / \phi_{k,g}'' \quad (\text{A. 16})$$

22 where the subscript ( $n\_UVn^0$ ) represents the initial values of UVn flash, and superscript (") indicates  
 23 the non-equilibrium values obtained from the flow-solver. Notably, the TPn flash is not only used in  
 24 the inner loop of UVn flash to compute the phase compositions, but also applied in the initialization to  
 25 calculate the initial equilibrium phase compositions ( $x_k, y_k$ ) (see Step (3) in Section 2.3).

26

### 27 A.3.2 Outer loop

28 In the outer loop, the specific internal energy and density of each phase are calculated by updating the  
 29 dependent variables of temperature and pressure with the phase compositions obtained from the TPn  
 30 flash. The objective functions are expressed as:

$$31 \quad F_3 = \frac{e^* - \sum_{p=1}^2 \psi_p M_p e_p / \bar{M}}{e^*} \quad (\text{A. 17})$$

$$32 \quad F_4 = \frac{\rho^* - \bar{M} / \sum_{p=1}^2 \psi_p M_p / \rho_p}{\rho^*} \quad (\text{A. 18})$$

33 The outer loop is performed using the initial estimates ( $P_{n\_UVn^0}$  and  $T_{n\_UVn^0}$ ) and a Newton iterative  
 34 algorithm (Saha and Carrolls, 1997). The internal energy and density of each phase ( $e_p$  and  $\rho_p$ ) are  
 35 computed based on the temperature derivatives of internal energy and density (Eqs. (A. 3) and (A. 5)),  
 36 as well as the pressure derivatives of density (Eq. (A. 2)). The first-order Taylor series approximation



1 with the relaxation coefficient ( $f_{rela} = 1 - \Delta T/T$ ) is used to estimate the temperature and pressure  
 2 variations ( $\Delta T$  and  $\Delta P$ ) as

$$3 \quad \Delta T = \frac{e^* - \sum_{p=1}^2 \psi_p M_p e_p / \bar{M}}{\partial e / \partial T}, \quad \Delta P = \Delta T \left. \frac{\partial V}{\partial T} \right|_P \left. \frac{\partial P}{\partial V} \right|_T \quad (\text{A. 19})$$

4 Therefore, the current pressure and temperature ( $P$  and  $T$ ) can be updated by  $P = P + f_{rela} \Delta P$  and  
 5  $T = T + f_{rela} \Delta T$ , respectively. The calculation of UVn flash is stopped if  $(F_3^2 + F_4^2) \leq \varepsilon_{UVn}$ .  
 6 Otherwise, the TPD stability test is applied again to determine the phase stability: if TPD = 2, update  
 7 the pressure and temperature ( $P$  and  $T$ ) again following Eq. (A. 19), and the TPn flash is iteratively  
 8 solved again with the new estimates for  $P$  and  $T$  until  $F_1 \leq \varepsilon_{UVn}$  and  $F_2 \leq \varepsilon_{UVn}$ ; if TPD = 0 or 1, the  
 9 mixture is assumed to be stable, and only the pressure and temperature ( $P$  and  $T$ ) are updated based on  
 10 Eq. (A. 19) without the TPn flash. In the outer loop of UVn flash, the pressure and temperature ( $P$  and  
 11  $T$ ) are initialized by the solutions at equilibrium state obtained from the UVn flash in the previous  
 12 time-step (denoted  $n - 1$ ) as

$$13 \quad P_{n\_UVn^0} = P_{n-1}, T_{n\_UVn^0} = T_{n-1} \quad (\text{A. 20})$$

14 Note that for  $n = 1$ , the input initial values  $P_0$  and  $T_0$  are used.

#### 16 A.4 Analytical solution of Cubic equation

17 There are three roots when solving the cubic EoS (e.g., PR EoS). In this study, an exact analytical  
 18 solution of cubic EoS is adopted based on the approach of (Perry, 1950, Wilczek - Vera and Vera,  
 19 2015). During the calculation, the non-physical meaning roots like negative or conjugate complex  
 20 values will appear. However, these non-physical roots are excluded, and only real positive roots are  
 21 selected. The detailed analytical solution is described as following:

$$22 \quad x^3 + Ax^2 + Bx + C = 0 \quad (\text{A. 21})$$

23 where,  $A, B$  and  $C$  are numerical coefficients.

25 Firstly, two coefficients,  $D$  and  $E$ , are defined as:

$$26 \quad D = \left(\frac{A}{3}\right)^3 - \left(\frac{AB}{6}\right) + \frac{C}{2} \quad (\text{A. 22})$$

$$27 \quad E = \left(\frac{B}{3}\right) - \left(\frac{A}{3}\right)^2 \quad (\text{A. 23})$$

29 Then, the discriminant is computed as  $\Delta = D^2 + E^2$ .

31 (1) If  $\Delta = 0$ , there are three roots with at least two equal roots as:

$$32 \quad x_1 = 2\sqrt[3]{-D} - \left(\frac{A}{3}\right), \quad x_2 = x_3 = -\sqrt[3]{-D} - \left(\frac{A}{3}\right) \quad (\text{A. 24})$$

33 (2) If  $\Delta > 0$ , there are one real root and two complex conjugate roots. Two other coefficients  $F$ ,  $G$  are  
 34 defined as:

1 
$$F = \sqrt[3]{(-D) + \sqrt{\Delta}}, G = \sqrt[3]{(-D) - \sqrt{\Delta}} \quad (\text{A. 25})$$

2 The roots are formulated as:

3 
$$\begin{cases} x_1 = F + G - \left(\frac{A}{3}\right) \\ x_2 = -\left[\frac{1}{2}(F + G) + \frac{A}{3}\right] + \frac{\sqrt{3}}{2}(F - G)i \\ x_3 = -\left[\frac{1}{2}(F + G) + \frac{A}{3}\right] - \frac{\sqrt{3}}{2}(F - G)i \end{cases} \quad (\text{A. 26})$$

4 (3) If  $\Delta < 0$ , there are three real and unequal roots. A new parameter  $\theta$  is defined as:

5 
$$\theta(\text{radians}) = \arccos\left(\frac{-D}{\sqrt{-E^3}}\right) \quad (\text{A. 27})$$

6 The three roots are formulated as:

7 
$$\begin{cases} x_1 = 2\sqrt{-E} \cos\left(\frac{\theta}{3}\right) - \frac{A}{3} \\ x_2 = 2\sqrt{-E} \cos\left(\frac{\theta}{3} + \frac{2}{3}\pi\right) - \frac{A}{3} \\ x_3 = 2\sqrt{-E} \cos\left(\frac{\theta}{3} + \frac{4}{3}\pi\right) - \frac{A}{3} \end{cases} \quad (\text{A. 28})$$

8

1 Reference

- 2 Allaire, G., S. Clerc, S. Kokh, 2002. A five-equation model for the simulation of interfaces between  
3 compressible fluids. *Journal of Computational Physics* 181, 577-616.
- 4 Aly, F.A., L.L. Lee, 1981. Self-consistent equations for calculating the ideal gas heat capacity, enthalpy, and  
5 entropy. *Fluid Phase Equilibria* 6, 169-179.
- 6 Amsden, A.A., 1997. KIVA-3V: A block-structured KIVA program for engines with vertical or canted  
7 valves. Los Alamos National Lab., NM, United States.
- 8 Amsden, A.A., P. O'rourke, T. Butler, KIVA-II: A computer program for chemically reactive flows with sprays.  
9 1989, Los Alamos National Lab., NM (USA).
- 10 Andrianov, N., R. Saurel, G. Warnecke, 2003. A simple method for compressible multiphase mixtures and  
11 interfaces. *International Journal for Numerical Methods in Fluids* 41, 109-131.
- 12 Baer, M., J. Nunziato, 1986. A two-phase mixture theory for the deflagration-to-detonation transition (DDT) in  
13 reactive granular materials. *International Journal of Multiphase Flow* 12, 861-889.
- 14 Baled, H., R.M. Enick, Y. Wu, M.A. McHugh, W. Burgess, D. Tapriyal, B.D. Morreale, 2012. Prediction of  
15 hydrocarbon densities at extreme conditions using volume-translated SRK and PR equations of state fit  
16 to high temperature, high pressure PVT data. *Fluid Phase Equilibria* 317, 65-76.
- 17 Banuti, D., 2015. Crossing the Widom-line–Supercritical pseudo-boiling. *The Journal of Supercritical Fluids* 98,  
18 12-16.
- 19 Battistoni, M., S. Som, D.E. Longman, 2014. Comparison of mixture and multifluid models for in-nozzle  
20 cavitation prediction. *Journal of Engineering for Gas Turbines and Power* 136, 061506.
- 21 Beig, S.A., E. Johnsen, 2015. Maintaining interface equilibrium conditions in compressible multiphase flows  
22 using interface capturing. *Journal of Computational Physics* 302, 548-566.
- 23 Bohbot, J., N. Gillet, A. Benkenida, 2009. IFP-C3D: an unstructured parallel solver for reactive compressible gas  
24 flow with spray. *Oil & Gas Science and Technology-Revue de l'IFP* 64, 309-335.
- 25 Castier, M., 2009. Solution of the isochoric–isoenergetic flash problem by direct entropy maximization. *Fluid  
26 Phase Equilibria* 276, 7-17.
- 27 Chiapolino, A., P. Boivin, R. Saurel, 2017. A simple and fast phase transition relaxation solver for compressible  
28 multicomponent two-phase flows. *Computers & Fluids* 150, 31-45.
- 29 Chung, T.H., M. Ajlan, L.L. Lee, K.E. Starling, 1988. Generalized multiparameter correlation for nonpolar and  
30 polar fluid transport properties. *Industrial & Engineering Chemistry Research* 27, 671-679.
- 31 Courant, R., K.O. Friedrichs, 1999. *Supersonic flow and shock waves*. Springer Science & Business  
32 Media.
- 33 Espósito, R.O., M. Castier, F.W. Tavares, 2000. Calculations of thermodynamic equilibrium in systems subject  
34 to gravitational fields. *Chemical Engineering Science* 55, 3495-3504.
- 35 Flåtten, T., H. Lund, 2011. Relaxation two-phase flow models and the subcharacteristic condition. *Mathematical  
36 Models and Methods in Applied Sciences* 21, 2379-2407.

1 Goncalvès, E., B. Charrière, 2014. Modelling for isothermal cavitation with a four-equation model. *International*  
2 *Journal of Multiphase Flow* 59, 54-72.

3 Habchi, C., 2015. A gibbs energy relaxation (GERM) model for cavitation simulation. *Atomization and Sprays*  
4 25,

5 Habchi, C., N. Dumont, O. Simonin, 2008. Multidimensional simulation of cavitating flows in diesel injectors by  
6 a homogeneous mixture modeling approach. *Atomization and sprays* 18,

7 Kapila, A., R. Menikoff, J. Bdzil, S. Son, D.S. Stewart, 2001. Two-phase modeling of deflagration-to-detonation  
8 transition in granular materials: Reduced equations. *Physics of fluids* 13, 3002-3024.

9 Kuijpers, M., D. Van Eck, M. Kemmere, J. Keurentjes, 2002. Cavitation-induced reactions in high-pressure  
10 carbon dioxide. *Science* 298, 1969-1971.

11 Kunz, R.F., D.A. Boger, D.R. Stinebring, T.S. Chyczewski, J.W. Lindau, H.J. Gibeling, S. Venkateswaran, T.  
12 Govindan, 2000. A preconditioned Navier–Stokes method for two-phase flows with application to  
13 cavitation prediction. *Computers & Fluids* 29, 849-875.

14 Kurschat, T., H. Chaves, G. Meier, 1992. Complete adiabatic evaporation of highly superheated liquid jets.  
15 *Journal of Fluid Mechanics* 236, 43-59.

16 Kwak, T., G. Mansoori, 1986. Van der Waals mixing rules for cubic equations of state. Applications for  
17 supercritical fluid extraction modelling. *Chemical Engineering Science* 41, 1303-1309.

18 Le Martelot, S., R. Saurel, B. Nkonga, 2014. Towards the direct numerical simulation of nucleate boiling flows.  
19 *International Journal of Multiphase Flow* 66, 62-78.

20 Le Métayer, O., R. Saurel, 2016. The Noble-Abel Stiffened-Gas equation of state. *Physics of Fluids* 28, 046102.

21 Linstrom, P.J., W. Mallard, 2001. NIST Chemistry webbook; NIST standard reference database No. 69.

22 Lund, H., 2012. A hierarchy of relaxation models for two-phase flow. *SIAM Journal on Applied Mathematics* 72,  
23 1713-1741.

24 Ma, P.C., Y. Lv, M. Ihme, 2017. An entropy-stable hybrid scheme for simulations of transcritical real-fluid  
25 flows. *Journal of Computational Physics* 340, 330-357.

26 Matheis, J., S. Hickel, 2018. Multi-component vapor-liquid equilibrium model for LES of high-pressure fuel  
27 injection and application to ECN Spray A. *International Journal of Multiphase Flow* 99, 294-311.

28 Menikoff, R., B.J. Plohr, 1989. The Riemann problem for fluid flow of real materials. *Reviews of modern*  
29 *physics* 61, 75.

30 Menter, F.R., 1994. Two-equation eddy-viscosity turbulence models for engineering applications. *AIAA journal*  
31 32, 1598-1605.

32 Michel, J., J. Franc, *Fundamentals of cavitation*. 2004, Springer.

33 Michelsen, M.L., 1982. The isothermal flash problem. Part I. Stability. *Fluid Phase Equilibria* 9, 1-19.

34 Moreau, J., O. Simonin, C. Habchi, 2004. A numerical study of cavitation influence on diesel jet atomisation.  
35 19th annual meeting of the institute for liquid atomization and spray systems, Nottingham, England,

36 Murrone, A., H. Guillard, 2005. A five equation reduced model for compressible two phase flow problems.  
37 *Journal of Computational Physics* 202, 664-698.

- 1 Pantano, C., R. Saurel, T. Schmitt, 2017. An oscillation free shock-capturing method for compressible van der  
2 Waals supercritical fluid flows. *Journal of Computational Physics* 335, 780-811.
- 3 Patankar, S., 1980. *Numerical heat transfer and fluid flow*. CRC press.
- 4 Peng, D.-Y., D.B. Robinson, 1976. A new two-constant equation of state. *Industrial & Engineering Chemistry*  
5 *Fundamentals* 15, 59-64.
- 6 Perry, J.H., *Chemical engineers' handbook*. 1950, ACS Publications.
- 7 Petitpas, F., E. Franquet, R. Saurel, O. Le Métayer, 2007. A relaxation-projection method for compressible flows.  
8 Part II: Artificial heat exchanges for multiphase shocks. *Journal of Computational Physics* 225, 2214-  
9 2248.
- 10 Petitpas, F., J. Massoni, R. Saurel, E. Lapebie, L. Munier, 2009. Diffuse interface model for high speed  
11 cavitating underwater systems. *International Journal of Multiphase Flow* 35, 747-759.
- 12 Qiu, L., Y. Wang, Q. Jiao, H. Wang, R.D. Reitz, 2014. Development of a thermodynamically consistent, robust  
13 and efficient phase equilibrium solver and its validations. *Fuel* 115, 1-16.
- 14 Ransom, V.H., D.L. Hicks, 1984. Hyperbolic two-pressure models for two-phase flow. *Journal of Computational*  
15 *Physics* 53, 124-151.
- 16 Saha, S., J.J. Carrolls, 1997. The isoenergetic-isochoric flash. *Fluid Phase Equilibria* 138, 23-41.
- 17 Saurel, R., R. Abgrall, 1999. A multiphase Godunov method for compressible multifluid and multiphase flows.  
18 *Journal of Computational Physics* 150, 425-467.
- 19 Saurel, R., P. Boivin, O. Le Métayer, 2016. A general formulation for cavitating, boiling and evaporating flows.  
20 *Computers & Fluids* 128, 53-64.
- 21 Saurel, R., A. Chinnayya, Q. Carmouze, 2017. Modelling compressible dense and dilute two-phase flows.  
22 *Physics of Fluids* 29, 063301.
- 23 Saurel, R., F. Petitpas, R. Abgrall, 2008. Modelling phase transition in metastable liquids: application to  
24 cavitating and flashing flows. *Journal of Fluid Mechanics* 607, 313-350.
- 25 Saurel, R., F. Petitpas, R.A. Berry, 2009. Simple and efficient relaxation methods for interfaces separating  
26 compressible fluids, cavitating flows and shocks in multiphase mixtures. *Journal of Computational*  
27 *Physics* 228, 1678-1712.
- 28 Schmitt, T., L. Selle, B. Cuenot, T. Poinso, 2009. Large-Eddy Simulation of transcritical flows. *Comptes*  
29 *Rendus Mécanique* 337, 528-538.
- 30 Segal, C., S. Polikhov, 2008. Subcritical to supercritical mixing. *Physics of Fluids* 20, 052101.
- 31 Simões-Moreira, J.R., 2000. Oblique evaporation waves. *Shock waves* 10, 229-234.
- 32 Simões-Moreira, J.R., J.E. Shepherd, 1999. Evaporation waves in superheated dodecane. *Journal of Fluid*  
33 *Mechanics* 382, 63-86.
- 34 Soave, G., 1972. Equilibrium constants from a modified Redlich-Kwong equation of state. *Chemical*  
35 *Engineering Science* 27, 1197-1203.

1 Tapriyal, D., R. Enick, M. McHugh, I. Gamwo, B. Morreale, High Temperature, high pressure equation of state  
2 density correlations and viscosity correlations. 2012, National Energy Technology Lab.(NETL),  
3 Pittsburgh, PA, and Morgantown, WV ....

4 Terashima, H., S. Kawai, M. Koshi, 2013. Consistent numerical diffusion terms for simulating compressible  
5 multicomponent flows. *Computers & Fluids* 88, 484-495.

6 Terashima, H., M. Koshi, 2012. Approach for simulating gas-liquid-like flows under supercritical pressures  
7 using a high-order central differencing scheme. *Journal of Computational Physics* 231, 6907-6923.

8 Thompson, P.A., H. Craves, G. Meier, Y.-G. Kim, H.-D. Speckmann, 1987. Wave splitting in a fluid of large  
9 heat capacity. *Journal of Fluid Mechanics* 185, 385-414.

10 Troshko, A., Y. Hassan, 2001. A two-equation turbulence model of turbulent bubbly flows. *International Journal*  
11 *of Multiphase Flow* 27, 1965-2000.

12 Utturkar, Y., J. Wu, G. Wang, W. Shyy, 2005. Recent progress in modeling of cryogenic cavitation for liquid  
13 rocket propulsion. *Progress in Aerospace Sciences* 41, 558-608.

14 Venkateswaran, S., J.W. Lindau, R.F. Kunz, C.L. Merkle, 2002. Computation of multiphase mixture flows with  
15 compressibility effects. *Journal of Computational Physics* 180, 54-77.

16 Vidal, J., 2003. *Thermodynamics: Applications in chemical engineering and the petroleum industry.*  
17 Editions Technip.

18 Wang, Y., L. Qiu, R.D. Reitz, R. Diwakar, 2014. Simulating cavitating liquid jets using a compressible and  
19 equilibrium two-phase flow solver. *International Journal of Multiphase Flow* 63, 52-67.

20 Wareing, C.J., R.M. Woolley, M. Fairweather, S.A. Falle, 2013. A composite equation of state for the modeling  
21 of sonic carbon dioxide jets in carbon capture and storage scenarios. *AIChE Journal* 59, 3928-3942.

22 Whitson, C.H., M.L. Michelsen, 1989. The negative flash. *Fluid Phase Equilibria* 53, 51-71.

23 Wilczek - Vera, G., J.H. Vera, 2015. Understanding cubic equations of state: A search for the hidden clues of  
24 their success. *AIChE Journal* 61, 2824-2831.

25 Wood, A., 1930. *A Textbook of Sound.* 1930. G. Bell and Sons Ltd., London 23.

26 Yang, S., C. Habchi, P. Yi, R. Lugo, Towards a multicomponent real-fluid fully compressible two-phase flow  
27 model, in ICLASS 2018, 14th Triennial International Conference on Liquid Atomization and Spray  
28 Systems. 2018: Chicago, IL, USA.

29 Yang, S., Y. Li, X. Wang, U. Unnikrishnan, V. Yang, W. Sun, 2017. Comparison of tabulation and correlated  
30 dynamic evaluation of real fluid properties for supercritical mixing. 53rd AIAA/SAE/ASEE Joint  
31 Propulsion Conference,

32 Zein, A., M. Hantke, G. Warnecke, 2010. Modeling phase transition for compressible two-phase flows applied to  
33 metastable liquids. *Journal of Computational Physics* 229, 2964-2998.

34