Modeling of non-equilibrium effects in intermittency region between two phases

Tomasz Waclawczyk

Warsaw University of Technology, Institute of Aeronautics and Applied Mechanics,
Division of Aerodynamics
Nowowiejska 24, 00653 Warszawa, Poland

Abstract

This paper concerns modeling of the evolution of intermittency region between two weakly miscible phases due to temporal and spatial variations of its characteristic length scale. First, the need of a more general description allowing for the evolution of intermittency region is rationalized. Afterwards, results of the previous work (Waclawczyk T., 2017, On a relation between the volume of fluid, level-set and phase field interface models, Int. J. Multiphas. Flow, Vol. 97) are discussed in context of the sharp interface models known in the literature and insight into droplet coalescence mechanism recently recognized in the molecular dynamics studies (Perumanath S., Borg M.K., Chubynsky M.V., Sprittles J.E., Reese J.M., 2019, Droplet coalescence is initiated by thermal motion, Phys. Rev. Lett., Vol. 122). Finally, the physical and numerical models extending applicability of the equilibrium solution to the case when intermittency region could also be in the non-equilibrium state is introduced and verified in several test cases.

Keywords: turbulent two-phase flow, non-equilibrium diffusive interface model, intermittency region, variable characteristic length-scale

*Tomasz Waclawczyk
Email address: tomasz.waclawczyk@pw.edu.pl (Tomasz Waclawczyk)
1. Introduction

The gas-liquid interface is a domain where the material properties of the two adjacent phases are changing. However, "the exact definition of the gas-liquid interface is nebulous" (Faust, 2018). In the fluid dynamics, there are two accepted physical models of the gas-liquid interface, namely: the dividing surface model (Gibbs, 1874) and the diffusive interface model (van der Waals, 1979). In the recent review paper (Elghobashi, 2019) concerning the direct numerical simulation (DNS) of the turbulent, dispersed two-phase flows, the numerical methods inspired by these two physical models of the gas-liquid interface are listed as "the tracking scalar approach". The dividing surface model of Gibbs is the foundation of the volume of fluid (VOF) (Tryggvason et al., 2011; Lu and Tryggvason, 2018) and standard level-set (SLS) (Osher and Sethian, 1988; Osher and Fedkiw, 2003; Sussman et al., 2007; Deike et al., 2016) sharp interface models. The diffusive interface model of van der Waals stimulated development of the phase-field methods based on the Cahn-Hilliard (Cahn and Hilliard, 1958; Anderson et al., 1998; Komrakova et al., 2015; Fedeli, 2017; Soligo et al., 2019) and Allen-Cahn equations (Allen and Cahn, 1979; Olsson and Kreiss, 2005; Chiu and Lin, 2011; McCaslin and Desjardins, 2014; Waclawczyk, 2015; Gruszczyński et al., 2020; Kajzer and Pozorski, 2020).

The common feature of the aforementioned physical and numerical models is the assumption that gas-liquid interface is an evolving in the turbulent velocity field boundary (geometric object) separating gas-liquid phases. As a consequence, to satisfy postulates of the DNS, the velocity field has to be resolved to the Kolmogrov length scale $\sim Re^{-3/4}$ to reconstruct all time and length scales governing evolution of this boundary. However, millions of droplets or bubbles created in the effect of violent topological changes can easily have the diameter below the Kolmogrov length scale (Elghobashi, 2019). To model their sub-grid dynamics and its impact on the flow field the phenomenological models are used.

To increase the range of Reynolds numbers, where the numerical simulations can offer useful predictions, some reduced models are obtained in the course of
filtering or ensemble averaging of the two-phase flow governing equations. These operations result, respectively, in the large-eddy (LES) (Labourasse et al., 2007; Toutant et al., 2007; Aniszewski et al., 2012; Herrmann, 2013; Saeedipour and Schneiderbauer, 2019) and Reynolds averaged (RANS) (Hong and Walker, 2000; Guo and Shen, 2010) formulations of the one-fluid model. After filtering or ensemble averaging of the one-fluid model equations, phenomenological models (often based on the DNS) are used to close correlations between the instantaneous (sub-grid) macroscopic interface and turbulent-velocity field (sub-grid) fluctuations. Regardless the reduction in the number of degrees of freedom, the gas-liquid interface in filtered/averaged one-fluid model is approximated in the same way as in the DNS. The characteristic scalar function defining the gas-liquid interface is transported using filtered or ensemble averaged fluid velocity. Hence again, the gas-liquid interface is viewed as the passive boundary between gas-liquid phases.

Thus, the gas-liquid interface model in the DNS, LES or RANS formulations of the one-fluid model does not play an active role in the modeling process. Additionally one notes, the aforementioned phenomenological models are often based on a different modeling strategy than this used in the one-fluid model, e.g. two-fluid or Euler-Lagrange frameworks (Prosperetti and Tryggvason, 2007; Elghobashi, 2019). This introduces coupling and feedback problems that must be addressed during the time-consuming simulations.

Recently the present author (Waclawczyk, 2017) has shown, the mathematical models describing the gas-liquid interface listed by (Elghobashi, 2019) as "the tracking scalar approach" are the complementary components of the gas-liquid interface statistical description. Herein, this result is extended and used to propose the modeling framework that is natural for the one-fluid model of two-phase flow.

In the present work it is assumed, the macroscopic intermittency region is a domain where the gas-liquid interface $\Gamma$ can be found with non-zero probability. This description was first introduced for the modeling of turbulence/gas-liquid interface interactions (Brocchini and Peregrine, 2001a,b). Therein, the sharp in-
terface $\Gamma$ is the gas-liquid interface, its ensemble averaged oscillations create the macroscopic intermittency region evolving due to stochastic, unsteady nature of turbulent flow. In this interpretation, interface $\Gamma$ deformations are caused by the stochastic forcing of the turbulent eddies, described typically in terms of the characteristic time and length scales, altered by the gravity and surface tension forces. The phenomenological model of Brocchini and Peregrine was used by several authors (Hong and Walker, 2000; Smolentsev and Miraghaie, 2005; Höhne and Vallée, 2009; Waclawczyk and Oberlack, 2011; Skartlien et al., 2014) to propose quantitative models of turbulence/gas-liquid interface interactions. In particular, Waclawczyk and Oberlack have proposed the correlation between local interface $\Gamma$ position and velocity fluctuation in the normal direction $\mathbf{n}_\Gamma$ must be modeled to account for evolution of the intermittency region. This idea was used to analyze the evolution of intermittency region based on a priori study of turbulent velocity field in vicinity of the sharp interface (Waclawczyk et al., 2014; Waclawczyk and Waclawczyk, 2015). Therein, it was found that the characteristic time $\tau_h \sim \epsilon_h/C [s]$ and length $\epsilon_h \sim D/C [m]$ scales are not constant but vary in time and space. The subject of present work is proposal of the model accounting for these characteristics of the macroscopic intermittency region.

Next, the present author (Waclawczyk, 2017) has shown, the intermittency region paradigm can be used to derive the equilibrium condition for the non-flat, gas-liquid interface $\gamma$ (mesoscopic intermittency region). The main argument therein is based on the analogy between processes of turbulence/gas-liquid interface and thermal-fluctuations/mesoscopic interface interactions. As it has been argued by (Brocchini and Peregrine, 2001a,b) and recently has been confirmed in the molecular dynamics studies by (Perumanath et al., 2019) both processes are stochastic in their nature. This means, derivation of the macroscopic (averaged) equations governing their evolution requires the conditional averaging taking into account instantaneous position of the sharp interface $\Gamma$. Moreover, one expects in the limit of vanishing energy of turbulent or thermal fluctuations, the more general, statistical model of intermittency region should
reduce to sharp interface models known in the literature.

Figure 1: Sketch of one realization of the considered stochastic process. The sharp interface \( \Gamma, \Psi(x,t) = 0 \) disturbed by the field of stochastic forces is oscillating around the expected position \( \psi(x,t) = 0 \) defining regularized interface \( \gamma \). Note, the intensity of \( \Gamma \) oscillations can in general be variable in space and/or time.

In the present work, the analogy between turbulence/gas-liquid interface and thermal-fluctuations/mesoscopic interface interactions is further exploited. We note, when the gas-liquid interface is in the equilibrium state, the characteristic length scale \( \epsilon_h(x,t) \) [m] governing its thickness is constant in time and space. In the opposite case the gas-liquid interface is in the non-equilibrium state. This scheme is extended to the case of turbulence/gas-liquid interface interactions. The distinction between equilibrium and non-equilibrium states of the macroscopic intermittency region, permits to address the question: to what extent classical sharp/diffusive interface models account for stochastic characteristics of the gas-liquid interface and why they allow predictions of topological changes governed by the molecular effects (Perumanath et al., 2019). Further, the physical and numerical models allowing to account for variable in space and time \( \epsilon_h(x,t) \) are proposed and used during numerical solution of the intermittency region evolution equation.

The present paper is organized as follows. In Section 2, equation governing evolution of the intermittency region is derived from the stochastic viewpoint.
and conditions of the intermittency region equilibrium and non-equilibrium are
defined. Afterwards, it is argued why the sharp/diffusive models of the inter-
mittency region have the potential to predict topological changes during break
up or coalescence occurring on a molecular level. In Section 3 it is shown, the
stationary solution of the intermittency region evolution equation accounting
for variable $\epsilon_h(x, t)$ is equivalent to minimization of the corresponding energy
functional. This result permits to use the stationary solution of the intermit-
tency region evolution equation as the equilibrium condition. In Section 3.2,
the equilibrium condition accounting for $\epsilon_h(x, t)$ is used to derive the mapping
function that accounts for the non-equilibrium effects. The generalized mapping
function is used during numerical solution of the intermittency region evolution
equation. In Section 4, the numerical method allowing to integrate the intermit-
tency region evolution equation is described and results of the intermittency
region evolving due to $\epsilon_h(x, t)$ are presented and discussed. In Section 5, con-
clusions and perspectives for future work are given.

2. Derivation of the intermittency region evolution equation

Let assume, the evolution of mesoscopic sharp interface $\Gamma$ is governed by the
phase indicator function $H_\Gamma(\Psi(x, t))$ transport equation

$$\frac{\partial H_\Gamma}{\partial t} + \mathbf{W} \cdot \nabla H_\Gamma = \frac{\partial H_\Gamma}{\partial t} + \delta_\Gamma(\Psi) |\nabla \Psi| \mathbf{W} \cdot \mathbf{n}_\Gamma = 0,$$

(1)

where $\Psi(x, t)$ [m] is the signed distance function from the points $\delta_\Gamma(\Psi)$ [1/m]
located on the two-dimensional surface $\Psi(x, t) = 0$ defining the sharp interface
$\Gamma$ with the normal vector $\mathbf{n}_\Gamma = \nabla \Psi / |\nabla \Psi|$, see Fig. 1. $\mathbf{W} \cdot \mathbf{n}_\Gamma$ [m/s] is stochastic
velocity field governing motion of $\delta_\Gamma(\Psi)$. $H_\Gamma(\Psi(x, t))$ represents one, instan-
taneous realization of the stochastic process generated by thermal fluctuations
$\mathbf{W}' = \mathbf{W} - \langle \mathbf{W} \rangle$ [m/s]. For this reason, Eq. (1) is of no use in the continuous
description of gas/fluid systems. To derive its continuum version the ensemble averag-
ing (Pope, 1998; Wacławczyk and Oberlack, 2011; Wacławczyk, 2017)
must be applied to Eq. (1). It is noticed, herein unlike in the recent work
(Thiesset et al., 2020) analysis is based on the one-point surface statistics. The ensemble average of the first LHS term in Eq. (1) results in

\[
\frac{\partial}{\partial t} \langle H_{\Gamma}(\Psi) \rangle = \frac{\partial}{\partial t} \int_{-\infty}^{\infty} H_{\Gamma}(\xi) \langle \delta(\Psi(x,t) - \xi) \rangle d\xi \tag{2}
\]

where \(\langle \delta(\Psi(x,t) - \xi) \rangle\) is the ensemble average \(\langle \cdot \rangle\) of the fine grained p.d.f.'s \(\delta(\Psi(x,t) - \xi)\) characterizing each realization of the stochastic process in the sample space \(\xi[m]\). \(\langle \delta(\Psi(x,t) - \xi) \rangle\) provides the probability density that \(\xi < \Psi(x,t) < \xi + d\xi\).

The contributions to the ensemble average \(\langle \cdot \rangle\) from the second RHS term in Eq. (1) are non-zero only if the interface \(\Gamma\) is present at point \(x\) and time \(t\) where/when the averaging is carried out. This issue was recognized by (Pope, 1998) who proposed the conditional surface average \(\langle \cdot \rangle_{\Gamma}\) to account for the smearing of the interface \(\Gamma\) due to the averaging process. Its application to the second RHS term in Eq. (1) leads to

\[
\langle W \cdot \nabla H_{\Gamma} \rangle = \langle W \cdot n_{\Gamma} \rangle_{\Gamma} \Sigma \tag{3}
\]

where \(\langle \cdot \rangle_{\Gamma}\) is the surface average

\[
\langle W \cdot n_{\Gamma} \rangle_{\Gamma} = \frac{1}{\Sigma} \int_{\Gamma} \langle W \cdot n_{\Gamma} \delta(\mu) \delta(\lambda) \delta(\Psi - \Psi'(\mu,\lambda,t)) A(\mu,\lambda,t) \rangle d\mu d\lambda \tag{4}
\]

and \(\Sigma[1/m]\) is given by the formula

\[
\Sigma = \int_{\Gamma} \langle \delta(\mu) \delta(\lambda) \delta(\Psi - \Psi'(\mu,\lambda,t)) A(\mu,\lambda,t) \rangle d\mu d\lambda. \tag{5}
\]

In equations (4-5) \(\mu, \lambda, \Psi\) define local, orthonormal coordinate system of the infinitesimally small element \(A(\mu,\lambda,t) d\mu d\lambda\) where \(\Psi\) is the coordinate in the normal direction. \(\Sigma[1/m]\) can be interpreted as amount of the expected surface-to-volume ratio (Pope, 1998); in the general case \(\Sigma(x,t)\) in Eq. (3) is unknown and must be closed by a model. Using decomposition \(W = \langle W \rangle + W'\) and Eqs. (3-5) one obtains

\[
\langle W \cdot n_{\Gamma} \rangle_{\Gamma} \Sigma = \langle W \rangle \langle n_{\Gamma} \rangle_{\Gamma} \Sigma + \langle W' \cdot n_{\Gamma} \rangle_{\Gamma} \Sigma. \tag{6}
\]

Next, the exact relations \(\langle W' \nabla H_{\Gamma} \rangle = \langle W' n_{\Gamma} \rangle_{\Gamma} \Sigma\) and \(\langle n_{\Gamma} \rangle_{\Gamma} \Sigma = \nabla \langle H_{\Gamma} \rangle\), see Eqs. (A.1) and (A.8) respectively, lead to Eq. (1) with the unclosed RHS correlation.
\[ \frac{\partial \alpha}{\partial t} + \mathbf{w} \nabla \alpha = -\langle \mathbf{W}' \cdot \mathbf{n}_\Gamma \rangle_{\Gamma \Sigma} = -\langle \mathbf{W}' \cdot \nabla H_\Gamma \rangle = -\langle \mathbf{W}' \cdot \mathbf{n}_\Gamma \delta_\Gamma (\Psi) \rangle, \quad (7) \]

where we have denoted \( \alpha = \langle H_\Gamma \rangle \) and \( \mathbf{w} = \langle \mathbf{W} \rangle \).

As it was put forward by (Waclawczyk and Oberlack, 2011), the unknown RHS term in Eq. (7) can be closed by the eddy diffusivity model

\[ \langle \mathbf{W}' \cdot \mathbf{n}_\Gamma \rangle_{\Gamma \Sigma} = -D \nabla \cdot \langle \mathbf{n}_\Gamma \rangle_{\Gamma \Sigma}. \quad (8) \]

Taking the divergence of exact relation \( \langle \mathbf{n}_\Gamma \rangle_{\Gamma \Sigma} = \nabla \alpha \) allows to derive

\[ \nabla \cdot \langle \mathbf{n}_\Gamma \rangle_{\Gamma \Sigma} = \nabla^2 \alpha - \langle \mathbf{n}_\Gamma \rangle_{\Gamma} \cdot \nabla \Sigma. \quad (9) \]

Substitution of Eq. (9) and Eq. (8) into Eq. (7) results in

\[ \frac{\partial \alpha}{\partial t} + \mathbf{w} \nabla \alpha = D \nabla^2 \alpha - D \langle \mathbf{n}_\Gamma \rangle_{\Gamma} \cdot \nabla \Sigma \quad (10) \]

where the second, unclosed RHS term in Eq. (10) was identified as the counter gradient diffusion. Above equation is not in the desired, conservative form. Thus, with the help of exact relation \( \langle \mathbf{n}_\Gamma \rangle_{\Gamma \Sigma} = \nabla \alpha \), taking into account the case when \( D(x, t) = C\epsilon_h(x, t) \) and noting the vector normal to the regularized interface \( \mathbf{n}_\gamma = \nabla \alpha / |\nabla \alpha| = \langle \mathbf{n}_\Gamma \rangle_{\Gamma} / |\langle \mathbf{n}_\Gamma \rangle_{\Gamma}| \), Eq. (10) may be rewritten as

\[ \frac{\partial \alpha}{\partial t} + \mathbf{w} \nabla \alpha = \nabla \cdot (D \nabla \alpha) - |\langle \mathbf{n}_\Gamma \rangle_{\Gamma}| \nabla (D \Sigma) \cdot \mathbf{n}_\gamma. \quad (11) \]

One notes, Eq. (11) accounts for the variable characteristic length scale \( \epsilon_h(x, t) \), however, it is still unclosed due to presence of the counter gradient diffusion term. The conservative closure of this unknown term (Waclawczyk and Waclawczyk, 2015) leads to equation first introduced by (Olsson and Kreiss, 2005) in the context of the conservative level-set (CLS) method

\[ \frac{\partial \alpha}{\partial t} + \nabla \cdot (\mathbf{w} \alpha) = \nabla \cdot [D \nabla \alpha |\mathbf{n}_\gamma - C\alpha (1 - \alpha) \mathbf{n}_\gamma] \quad (12) \]

where \( \mathbf{w}[m/s] \) is velocity of the regularized interface \( \gamma \), and in the general case \( C(x, t) [m/s] \) and \( D = C\epsilon_h(x, t) [m^2/s] \) are velocity and diffusivity scales characterizing the intermittency region, respectively. We note, the presence of two
RHS terms in Eq. (12) is supported by the fact that forces always occur in pairs: diffusion $D|\nabla \alpha|n_\gamma$ due to presence of the sharp $\alpha$ profile is counterbalanced by contraction $Ca (1 - \alpha) n_\gamma$. The latter term was identified as the first order approximation of joint probability of creation of the bond between particles of two different types (Cahn and Hilliard, 1958).

The steady state solution of Eq. (12) with $\epsilon_h = const.$ and $w = u = 0$ is given by the regularized Heaviside function

$$\alpha (\psi) = \frac{1}{1 + \exp (-\psi (x, t)/\epsilon_h)} = \frac{1}{2} \left[ 1 + \tanh \left( \frac{\psi (x, t)}{2\epsilon_h} \right) \right]$$

and its inverse function that is the signed distance from the expected position of the regularized interface $\gamma$ defined by the level-set $\psi (\alpha = 1/2) = 0$

$$\psi (\alpha) = \epsilon_h \ln \left[ \frac{\alpha (\psi)}{1 - \alpha (\psi)} \right].$$

As noticed by the present author (Wacławczyk, 2015), Eqs. (13) and (14) are known to characterize the cumulative distribution $\alpha (\psi)$, and quantile $\psi (\alpha)$ functions of the logistic distribution. Additionally, the gradient of $\alpha (\psi)$ given by the formula

$$\nabla \alpha = \frac{\delta (\alpha)}{\epsilon_h} \nabla \psi,$$

where $\delta (\alpha) / \epsilon_h = \alpha (1 - \alpha) / \epsilon_h$ is the probability density function of the logistic distribution. Eq. (15) allows to reformulate Eq. (12) to

$$\frac{\partial \alpha}{\partial t} + w \nabla \alpha = \nabla \cdot \left[ C\delta (\alpha) (|\nabla \psi| - 1) n_\gamma \right],$$

where $n_\gamma = \nabla \alpha / |\nabla \alpha| = \nabla \psi / |\nabla \psi|$, and $w$ in Eq. (14) may now be replaced by fluid velocity $u$ as in absence of phase changes and/or advection $n_\gamma \cdot w = 0$.

In the present work we separate advection and re-initialization steps in Eq. (16) what leads to

$$\frac{\partial \alpha}{\partial t} + w \nabla \alpha = \frac{\partial \alpha}{\partial t} + \frac{\delta (\alpha)}{\epsilon_h} w \cdot \nabla \psi = 0,$$

$$\frac{\partial \alpha}{\partial \tau} = \nabla \cdot \left[ C\delta (\alpha) (|\nabla \psi| - 1) n_\gamma \right].$$
This form of Eq. (12) is preferred as it allows to focus separately on advection Eq. (17) and model for the evolution of intermittency region using Eq. (18). Solution of Eq. (18), known in the literature as the re-initialization step, was shown to be equivalent to minimization of the interfacial energy functional containing the term which accounts for regularized interface $\gamma$ deformation. For this reason it was argued, Eqs. (17) and (18) with the mapping between $\alpha(\psi) - \psi(\alpha)$ functions given by Eq. (14) describe the non-flat, intermittency region in the equilibrium state as $C = \text{const.}$, $D = C\epsilon_h = \text{const.}$ in the previous work (Wacławczyk, 2017). Furthermore, the statistical interpretation of Eqs. (17) and (18) based on Eqs. (13)-(15) reveals the relation between the sharp and diffusive interface models.

As it was already mentioned in Section 1, Eq. (11) can be interpreted as the mesoscopic or macroscopic statistical model of the intermittency region depending upon character of the stochastic force field inducing $\mathbf{W}$ in Eq. (1) and chosen time/length scales. In the mesoscopic interpretation, deformation of the sharp interface $\Gamma$ is caused by the random, thermal fluctuations. In the macroscopic interpretation, velocity $\mathbf{W}$ in Eq. (1) can be related to the instantaneous turbulent velocity field. In the next section, terms under which the intermittency region is in the equilibrium or non-equilibrium state are discussed.

2.1. Equilibrium and non-equilibrium state of the intermittency region

First, the mesoscopic interface $\Gamma$ agitated by the thermal fluctuations is considered, see Fig. 1. After conditional averaging described in Section 2 its evolution is described by Eq. (12). Let us note, $\alpha(\psi)$ and $\delta(\alpha)$ in Eq. (12) have an infinite support as $\epsilon_h \sim \sqrt{k_B T/\sigma} > 0$ where $k_B [\text{J/K}]$ is the Boltzmann constant, $T [\text{K}]$ is absolute temperature and $\sigma [\text{J/m}^2]$ is the surface tension coefficient (Vrij, 1973; Aarts et al., 2004). $k_B T/V [\text{J/m}^3]$ is the root mean square measure of the thermal fluctuations of molecules acting to distort the mesoscopic interface $\Gamma$ between two phases in the infinitesimally small volume $V$. The amount of thermal energy in $V$ determines the kinetic energy of these molecules. A surface tension $\sigma/V [\text{J/m}^3]$ represents net work done by the cohesive forces
between fluid molecules per unit area of $\Gamma$ in $V$. The cohesive forces between fluid molecules act to suppress increases in interfacial area of $\Gamma$.

In the case of the turbulence/gas-liquid interface interactions, the interface $\Gamma$ in Fig. 1 is the sharp representation of the gas-liquid, macroscopic interface. By the analogy to the mesoscopic case, the characteristic length scale $\epsilon_h (x,t)$ is governed by the ratio of net turbulent kinetic energy $\rho k [J/m^3]$ and the work of forces generating turbulent stresses per unit area of the interface $\Gamma$ in $V$, $[J/m^5]$. These forces are acting to decrease or increase the interface $\Gamma$ area. In the macroscopic interpretation of Eq. (1), the ratio of works done by volume/surface forces in the intermittency region is altered by the works done by the gravitational and surface tension forces, respectively.

![Figure 2: Schematic picture of the intermittency region in (a) equilibrium $\epsilon_h = const.$ and (b) non-equilibrium $\epsilon_h (x,t)$ states. The expected position of the interface $\gamma$ is depicted using black solid line.](image)

If the ratio of works done by the random volume forces (inducing fluctuations of $\Gamma$) and surface forces (per unit area of $\Gamma$) is constant in time and space, then also $\epsilon_h (x,t) = const.$ and the intermittency region is in the equilibrium state, see Fig. 2a. In the opposite case, the characteristic length scale $\epsilon_h (x,t)$ may change in time and space, and for this reason, the intermittency region is in the non-equilibrium state, see Fig. 2b.

In what follows, the statistical interpretation of Eq. (12) is used to localize the sharp interface tracked or captured in the VOF, SLS sharp interface models.
In the series of molecular dynamics simulations of two droplets collisions (Perumanath et al., 2019) have identified characteristic thermal length scale \( l_T \sim 2\sqrt{\epsilon_h R} \) where \( \epsilon_h \sim \sqrt{k_B T / \sigma} \) is the intermittency region thickness, \( R \) denotes colliding droplets radii. The existence of the thermal length scale \( l_T \) shows, before the capillary forces take control on the droplets coalescence, molecular, thermal effects govern this process. Therefore, Perumanath et al. conclude the droplet coalescence is stochastic phenomenon initiated by thermal motion of fluid particles. Since \( l_T \) is proportional to the square root of the droplet radius \( R \), it is expected that during topological changes the molecular effects may influence phenomena on the macroscopic scale resolved in fluid dynamics.

In the light of this fact, classical sharp interface models seem to overlook molecular effects (see derivations the signed-distance and phase indicator function transport equations in (Osher and Fedkiw, 2003; Tryggvason et al., 2011), respectively). Hence, the natural question arises: to what extent the sharp interface models are able to reconstruct topological changes of the gas-liquid interface during coalescence and/or break up events?

First we note, the expected position of the regularized (macroscopic) interface \( \gamma: \psi = 0 \) is different than the instantaneous position of the mesoscopic sharp interface \( \Gamma: \Psi = 0 \) defining one realization of the stochastic process, see Fig. 1. Since in the case of gas-liquid intermittency region in the equilibrium state \( 0 < \epsilon_h < 1 \) and \( \epsilon_h = const. \), one can assume \( \epsilon_h \) does not depend on volume and surfaces forces performing works in the domain where two-phase system is changing its properties. Hence, \( \epsilon_h \to 0 \) means \( \alpha(\psi) \to H_\gamma (\psi = 0) = 1/2 \).

For this reason, similar to Gibbs dividing surface neglecting information about \( 0 < \epsilon_h < 1 \), the sharp interface models are valid only when it is assumed the intermittency region is in the equilibrium state.

The phase indicator function \( H_\gamma \) built on the expected position of the regularized interface \( \gamma: \psi(x, t) = 0 \) is different than the phase indicator function \( H_\Gamma \) built on the signed distance function \( \Psi(x, t) \) as \( H_\Gamma (\Psi(x, t)) \) is one realiza-
tion of the stochastic process governed by Eq. (1). $H_\gamma$ is the phase indicator function discretized in the VOF methods, $\psi(x,t)$ is the signed distance function discretized in the SLS methods. The level-sets $H_\gamma = 1/2, \psi = 0$ are two equivalent geometric, two-dimensional representations of the expected position of the gas-liquid regularized interface $\gamma$ as $0 < \epsilon_h < 1$ must remain greater than zero. This explains how molecular effects are taken into account in the VOF, SLS type sharp interface models.

Because the sharp interface approximations are formulated in the limit of $\epsilon_h \to 0$, they do not depend explicitly on $\epsilon_h$ and hence do not allow to account for the non-zero volume/interfacial energy ratio governing the intermittency region width, its possible variations and consequences of these, too. Thus, without additional modeling assumptions the VOF, SLS type sharp interface models can not account for thermal effects described by (Perumanath et al., 2019). However, due to the fact VOF, SLS interface models sharply reconstruct the expected position of the regularized gas-liquid interface $\gamma$: $H_\gamma = 1/2, \psi = 0$, respectively, they are able to approximate break up and (in most of the cases) coalescence processes.

As long as the intermittency region remains in the equilibrium state and/or energy of stochastic fluctuations is small and independent of background physical phenomena the sharp interface model is a good approximation. In the opposite case, some physical effects may be lost when using the sharp interface model as local variations of the volume/surface forces work ratio in the intermittency region can affect dynamics of adjacent gas-liquid phases. For example through the local modifications of their material properties. In the remaining part of the present paper it is proposed how description given by Eqs. (17, 18, 14) can be extended to model the intermittency region in the non-equilibrium state.

3. Modeling of non-equilibrium effects in the intermittency region

As it has been explained in the previous sections, the motivation for generalized numerical solution of Eqs. (17, 18, 14) comes from the need to account
for the case when the ratio of works performed by the volume/surface forces governing $\epsilon_h$ varies in space and time. Generalization of the equilibrium model can also justified from a thermodynamic perspective. The intermittency region between two weakly miscible phases is an open system that may not be in the equilibrium state as it is perpetually exchanging energy with neighboring phases.

During previous analytical considerations and numerical experiments it has been assumed $C = \text{const.}$ and $\epsilon_h \sim \Delta x$, see Fig. 2a. The main subject of the present section is to extend the analytical model and numerical solution of the set of differential algebraic Eqs. (17, 18, 14) to the case when $\epsilon_h(x, t)$ is variable as it is schematically depicted in Fig. 2b. The case when the characteristic time scale $\tau_h \sim \epsilon_h(x, t)/C(x, t)$ is variable too, is left for future studies.

3.1. Minimization of free energy functional with variable characteristic length scale

In the previous work of the present author (Wacławczyk, 2017) it has been shown the Helmholtz free energy functional defining the energy of two-phase system

$$F[\alpha] = \int_V \sigma \left[ \epsilon_h |\nabla \alpha|^2 + \frac{f(\alpha)}{\epsilon_h} + k(\alpha) \right] dV$$  (19)

where $\sigma [J/m^2]$ is a known constant and $f(\alpha) = \alpha^2 (1 - \alpha)^2 [-]$, has to contain term $k(\alpha)$ accounting for the energy of the interface $\gamma$ deformation. Its presence in Eq. (19) is required to guarantee the equilibrium state of the non-flat regularized interface $\gamma$ by setting $\delta F/\delta \alpha = 0$. From, the equilibrium condition given by the stationary solution to Eq. (12) with $C = \text{const.}, \epsilon_h = \text{const.}$ it was shown $k(\alpha)$ in Eq. (19) satisfies the relation

$$\int_V \frac{\partial k(\alpha)}{\partial \alpha} dV \delta \alpha = \int_V 2\alpha (1 - \alpha) \nabla \cdot \mathbf{n}_\gamma dV \delta \alpha.$$  (20)

As $\delta k/\delta \alpha$ in Eq. (20) does not depend explicitly on $\epsilon_h(x, t)$ above relation will also be used herein.

Next it is shown, the functional derivative of Eq. (19) with the variable characteristic length scale $\epsilon_h(x, t)$ leads to the stationary solution of Eq. (12)
accounting for the non-equilibrium effects. The RHS of stationary Eq. (12) leads to re-initialization equation in the non-conservative form

\[
\frac{\partial \alpha}{\partial \tau} = \epsilon_h \nabla^2 \alpha + \nabla \epsilon_h \cdot \nabla \alpha - \frac{\alpha (1-\alpha)}{\epsilon_h} \left[(1 - 2\alpha) + \epsilon_h \nabla \cdot n_\gamma \right] 
\]

(21)

where we set \( C = 1 \, [m/s] \) for clarity.

Calculation of the functional derivative of Eq. (19) with \( \epsilon_h(x,t) \) is carried out in Appendix B. The minimization condition given by Eq. (B.6) is the same as the RHS of Eq. (21), therefore

\[
\frac{\delta F}{\delta \alpha} = \frac{\partial \alpha}{\partial \tau} = 0. 
\]

(22)

The stationary solution to Eq. (12) or the steady state solution of the corresponding re-initialization equation in pseudo-time \( \tau \) would minimize the functional (19) with the variable characteristic length scale \( \epsilon_h(x,t) \). The additional term \( \nabla \epsilon_h \cdot \nabla \alpha \) forces changes of the \( \alpha \) function shape. When \( \epsilon_h = \text{const.} \) the equilibrium solution given by Eqs. (13) and (14) is recovered. In the following section, the mapping function used during numerical solution of Eqs. (17) and (18) is derived.

3.2. Modification of the mapping procedure

In the present section it is proposed how to use the re-initialization equation in the form of Eq. (18) taking into account variable \( \epsilon_h(x,t) \). The equilibrium condition obtained from stationary solution to Eq. (12) reads

\[
\nabla \alpha = |\nabla \alpha| n_\gamma = \frac{\alpha (1-\alpha)}{\epsilon_h(x,t)} n_\gamma. 
\]

(23)

Eq. (23) is formulated in the direction \( n_\gamma \) normal to the regularized interface \( \gamma \), hence it may be rewritten as

\[
\left. \frac{\partial \alpha}{\partial \psi} \right|_{\partial x} = \frac{1}{\epsilon_h(x,t)} \alpha (1-\alpha), 
\]

(24)

where it is assumed \( \partial \alpha/\partial \psi > 0 \) meaning \( \alpha (\psi) \) is expected to be the cumulative distribution function with the infinite support due to analogy with Eq. (15). Next, we assume \( |\nabla \psi| \equiv 1 \) in Eqs. (23) and (24). As a result, substitution
of Eq. (23) into Eq. (12) with \( D(x, t) = C\epsilon_h(x, t) \) allows to derive Eq. (18). The assumption \(|\nabla \psi| \equiv 1\) means, the signed distance function \( \psi(x, t) \) spans the space where surface averaged oscillations of the sharp interface \( \Gamma \) take place. On average, these oscillations occur only in the direction \( n_\gamma \) normal to the expected position \( \psi = 0 \) of the regularized interface \( \gamma \). Above interpretation explains the difference between \( \psi(\alpha) \) and \( \Psi(x, t) \) signed distance function fields. \( \Psi(x, t) \) is exclusively the signed distance from points \( \delta\Gamma(\Psi) \) located at the sharp interface \( \Gamma \) defined by the level-set \( \Psi(x, t) = 0 \).

Further, it is noticed at each point \((x, t)\) of the field \( \epsilon_h(x, t) \) the signed distance function \( \psi(x, t) \) is given. Hence, knowing \( \psi(x, t) \) allows to obtain \((x, t)\) and thus \( \epsilon_h(x, t) \). Therefore, we introduce \( \epsilon_{\psi}^h(x, t) \) denoting \( \epsilon_h(x, t) \) determined using \( \psi(x, t) \). Usage of \( \epsilon_{\psi}^h(x, t) \) allows to integrate Eq. (24) in the local coordinate system attached to the regularized interface \( \gamma \). As \( \gamma \) is defined by \( \psi(x, t) = 0 \), \( \psi(x, t) \) is the normal coordinate with the origin at \( \psi(x, t) = 0 \) of this local system. At each, fixed point of given \( \alpha(\psi), \psi(\alpha), \epsilon_h(x, t) \) fields this integration reads

\[
\int_{\alpha(\psi)}^{1/2} d\alpha' = \int_{\psi(\alpha)}^{0} \frac{d\psi'}{\epsilon_{\psi}^h(x, t)}.
\]

The integration (25) is performed from the arbitrary point located at the signed-distance from the regularized interface \( \alpha(\psi) - \psi(\alpha) \) to the expected position of the regularized interface \( \psi(\alpha = 1/2) = 0 \). One notes, the LHS integration in Eq. (25) does not assume or result in any specific form/shape of the function \( \alpha(\psi) \).

To recover the equilibrium solution when \( \epsilon_h(x, t) = const. \) it is necessary to preserve mapping between \( \alpha(\psi) - \psi(\alpha) \), see Eq. (14). For this reason, it is more convenient to reformulate the RHS integral in Eq. (25) using the variable substitution as follows

\[
\int_{\psi(\alpha)}^{0} \frac{d\psi'}{\epsilon_{\psi}^h(x, t)} = \psi(\alpha) \int_{1}^{0} \frac{dt'}{\epsilon_{\psi}^h(x, t)} = \psi(\alpha) I(\psi)
\]

where \( t' \in [0, 1] \) is the parameter such that \( \psi' = t'\psi \) and \( d\psi' = dt'\psi \), furthermore \( I(\psi) \) is used to denote integral on the RHS of Eq. (26). After integration of
Eq. (24) with Eq. (26) one obtains

$$\psi(\alpha) = \frac{1}{\mathcal{I}(\psi)} \ln \left[ \frac{\alpha(\psi)}{1 - \alpha(\psi)} \right].$$  \hspace{1cm} (27)

At the given, arbitrary point \((x, t)\), the signed distance \(\psi(x, t)\) has the known value. For this reason, at the point \((x, t)\) the integral \(\mathcal{I}(\psi) = \text{const.}\) and thus an inverse relation is also true

$$\alpha(\psi) = \frac{1}{1 + \exp(-\psi(\alpha)\mathcal{I}(\psi))}.\hspace{1cm} (28)$$

The only difference between Eqs. (14) and (13) and Eqs. (27) and (28) is the latter take into account variation of \(\epsilon_h(x, t)\) in the sense of Eq. (25). When the field \(\epsilon_h(x, t) = \text{const.}\), Eq. (25) and Eq. (26) reduce to the equilibrium solution used to derive Eqs. (13) and (14). Thus, the mapping given by Eq. (27) or Eq. (28) can be used during numerical solution of the system given by Eqs. (17) and (18) to model how the \(\epsilon_h(x, t)\) field is affecting changes of the cumulative distribution function \(0 < \alpha(\psi) < 1\) profile. When \(\epsilon_h(x, t) = \text{const.}\) the equilibrium solution is recovered by definition of \(\mathcal{I}(\psi)\).

4. Numerical solution

In this section a numerical method for the exact and approximate solutions of the intermittency region evolution equation with the structured grid solver is introduced. First, the one-dimensional study is carried out showing how the re-initialization equation (18) with the modified mapping procedure defined by Eq. (27) can be used to reconstruct the intermittency region in the non-equilibrium state. Afterwards, the coupled solution is compared with the semi-analytical approach using Eq. (18) where \(\epsilon_h = \text{const.}\) and Eq. (28) is accounting for variable \(\epsilon_h(x, t)\). Finally, the semi-analytical solution is used in two-dimensional studies without and with advection to reconstruct more complex behavior of the intermittency region. Details of discretization and numerical solution of Eqs. (17) and (18) using the mapping given by Eq. (14) where \(\epsilon_h = \text{const.}\) are described in (Waclawczyk, 2015, 2017). In Appendix D, minor modifications to these schemes required to take into account variable \(\epsilon_h(x, t)\) are given.
4.1. Finding quadrature

The main problem during numerical solution of the set of differential algebraic equations (17, 18, 27) or (17, 18, 14) and Eq. (28) is approximation of the integral (26). Namely, one needs to find the quadrature \( I(\psi) \) for integral

\[
I(\psi) = \int_{t_1}^{t_0} \frac{d\psi}{\epsilon_h(x,t)} \approx I(\psi)
\]

keeping in mind the parameter \( t' \) is changing along the signed distance function \( \psi(\alpha) \), from the local position on the computational grid at \( t=1 \) to the interface at \( t=0 \). As function \( \psi(\alpha) \) is also the solution to Eq. (18), one can use this to formulate the effective numerical integration procedure.

Figure 3: Determination of the interface \( \gamma \) position on structured grid using signed distance function \( \psi(\alpha) \) (not depicted). Schematic presentation of the points at \( t=0 \) (int) and \( t=1 \) (inp) required to compute the trapezoidal quadrature, see Eq. (30).

Let note, if the two point, first-order accurate quadrature is used the discussed problem is reduced to finding index (int) of the control volume with the interface \( \gamma \) during loop over all grid points with the index (inp). The sketch of this procedure in the two-dimensional case for the one pair of control volumes (inp) and (int) is depicted in Fig. 3. The simplest quadrature taking into account the two point information required to approximate Eq. (29) is given by
the first-order accurate trapezoidal rule

\[ I_T(\psi) = \frac{1}{2} \left[ \frac{1}{\epsilon_h(\text{int})} + \frac{1}{\epsilon_h(\text{inp})} \right]. \]  

(30)

To obtain higher order of accuracy, the third-order accurate Simpson rule can be used to approximate Eq. (29), it reads

\[ I_S(\psi) = \frac{1}{6} \left[ \frac{1}{\epsilon_h(\text{int})} + 4 \frac{1}{\epsilon_h(\text{inm})} + \frac{1}{\epsilon_h(\text{inp})} \right]. \]

(31)

where (inm) denotes control volume in the center between (inp), (int). In what follows it is compared how approximations of the integral (29) given by Eqs. (30) and (31) affect obtained solutions. Introduction of even higher order of accuracy in approximation of Eq. (26) requires considering additional control volumes in-between local position on the mesh (inp) and expected position of the interface \( \gamma \) (int). When the number of control volumes between (inp) and (int) is smaller than the quadrature stencil, the higher-order quadrature have to be replaced by the appropriate lower-order quadrature or interpolation of \( \epsilon_h(x,t) \).

As the present results are obtained in the code using structured grid solver, computation of Eqs. (30) and (31) is straightforward. Knowing local position at the grid (inp) and value of the signed distance function in this cell \( \psi(\text{inp}) \) one needs to project it on \((x,y)\) directions to obtain: \( \psi_x = -\psi(\text{inp} ) n_{\gamma,x} \), \( \psi_y = -\psi(\text{inp} ) n_{\gamma,y} \). Next, compute constants \((L,M)\) (see Fig. 3) where \( L \approx NINT(\psi_x/\Delta x) \) and \( M \approx NINT(\psi_y/\Delta y) \), and finally determine index of the cell containing interface (int), on structured grid \( \text{int} = \text{inp} + L \cdot NJ + M \), where \( NJ \) is number of grid cells in \( j \) direction, \( NINT \) is the intrinsic function returning nearest integer. Point (inm) in Eq. (31) is obtained in similar way taking \( \psi_x^{\text{m}} = -0.5\psi(\text{inp} ) n_{\gamma,x} \), \( \psi_y^{\text{m}} = -0.5\psi(\text{inp} ) n_{\gamma,y} \) and then computing \( (L^m, M^m) \). If the stencil where Eq. (31) is computed is smaller than three control volumes, the \( \epsilon_h(\text{inm}) \) value is obtained as \( \epsilon_h(\text{inm}) = (\epsilon_h(\text{inp}) + \epsilon_h(\text{int})) / 2 \). Due to introduction of \( \epsilon_h^{\prime}(x,t) \) in Eq. (25), in one and three dimensional cases this procedure can be easily adopted taking into account one less or one more spatial direction to compute (int) and/or (inm).
4.2. Evolution of one-dimensional cumulative distribution function

To compare the exact and approximate semi-analytical solution, evolution of the one-dimensional $\alpha(\psi)$ profile disturbed by variable $\epsilon_h(x,t)$ is considered. $\epsilon_h(x,t)$ is predefined as the step $\epsilon_{h,S}(x,t)$ or bell $\epsilon_{h,B}(x,t)$ shaped disturbance, see Eqs. (C.1) and (C.2) respectively. In both cases only Eq. (18) is solved, as $w = u = 0$ advection is neglected. The computational domain $[0,1]$ is divided into 128 control volumes, the number of physical time steps is set to $N_t = 72$, $\Delta t = 10^{-3}$, the number of re-initialization time steps is set to $N_\tau = 32$, with size $\Delta \tau = \epsilon_{h,b}/2$; the minimum (base) thickness of the interface is set to $\epsilon_{h,b} = \Delta x$. The discretization of Eq. (18) is the same as in (Wachawczyk, 2017), the only modification accounting for $\epsilon_h(x,t)$ is introduced to the constrained interpolation used to approximate $\tilde{\delta}(\alpha) = \alpha(1-\alpha)$ in Eq. (18), see Eq. (D.5).

Figs. 4 – 5 depict, respectively, evolution of $\epsilon_{h,S}(x,t)$, $\epsilon_{h,B}(x,t)$ profiles and corresponding variations of $\alpha(\psi)$ at equal time intervals. The black-dashed lines represent the analytical $\alpha(\psi)$ profiles obtained using Eq. (13) with $\epsilon_h = \epsilon_{h,b}$ (dashed-line), and $\epsilon_h = 2\epsilon_{h,b}$ (dashed-dotted line). In Figs. 4 – 5, $\alpha_{dc}$ denotes solutions obtained using the direct coupling of Eqs. (18) and (27), $\alpha_d$ denotes approximate, semi-analytical solution obtained using Eq. (18) where $\epsilon_h = \epsilon_{h,b}$ and Eq. (28) accounting for $\epsilon_h(x,t)$. In both cases the third-order accurate Simpson rule (31) is used to approximate $I(\psi)$ integral, see Eq. (26).

The convergence space of the re-initialization equation (18) for the coupled cases $\alpha_{dc}$ is presented in Fig. 6. Therein, the $L_{1,\tau}$ norm defined by Eq. (E.1) characterizing the numerical solution of Eq. (18) in times $t, \tau$ is presented. The top row presents convergence of Eq. (18) obtained using the first-order accurate quadrature (30), the bottom row using the third-order accurate quadrature (31). Although in all cases convergence of the numerical solution is obtained, one observes variation of $\alpha(\psi)$ caused by $\epsilon_h(x,t)$ strongly affects the numerical solution of Eq. (18). In order to avoid this dependence the case where $\epsilon_h = \epsilon_{h,b} = const.$ in Eq. (18) and $\epsilon_h(x,t)$ variation is modeled using Eq. (28) is considered. In this case, the $L_{1,\tau}$ norm remains almost constant $\sim 10^{-16}$ for all steps $t, \tau$; obtained $\psi$ is treated as the carrier function for $\alpha(\psi)$. 

20
Figure 4: The comparison of \(\alpha(\psi)\) affected by step shaped variation of \(\epsilon_{h,S}(x,t)\) given by Eq. (C.1) depicted after (left to right) equal time intervals. Black-dashed lines depict the analytical profiles of \(\alpha(\psi)\) where \(\epsilon_h = \epsilon_{h,b}\) and \(\epsilon_h = 2\epsilon_{h,b}\), see Eq. (13).

The differences between \(\alpha_{dc}\) and \(\alpha_d\) profiles observed in Figs. 4 – 5 are the consequence of differences in \(\psi\) fields obtained during the coupled and semi-analytical solutions. During the direct coupling, \(\psi(\alpha)\) is the part of numerical solution and hence, the expected position of the interface \(\psi = 0\) can change its location. In the semi-analytical case, \(\epsilon_h = \epsilon_{h,b} = \text{const.}\), for this reason the
The comparison of $\alpha(\psi)$ affected by bell shaped variation of $\epsilon_{h,B}(x,t)$ given by Eq. (C.2) depicted after (left to right) equal time intervals. Black-dashed lines depict the analytical profiles of $\alpha(\psi)$ with $\epsilon_h = \epsilon_{h,b}$ and $\epsilon_h = 2\epsilon_{h,b}$, see Eq. (13).

position $\psi=0$ is not affected by variations of the characteristic length scale field $\epsilon_h(x,t)$. The latter approach simplifies numerical solution of Eq. (18), but as it can be observed in Figs. 4 – 5, the semi-analytical solution $\alpha_d$ closely mimics the exact one $\alpha_{dc}$.

Fig. 7 illustrates, how the order of accuracy of the quadrature used in Eq.
Figure 6: Convergence space of $L_{1,\tau}$ norm given by Eq. (E.1), during solution of Eq. (18) and Eq. (27) obtained with $N_t = 72$ time and $N_{\tau} = 32$ re-initialization steps. The one-dimensional $\alpha(\psi)$ profile is altered by variable $\epsilon_{h,S}(x,t)$ a),c) or $\epsilon_{h,B}(x,t)$ b),d). Solutions are obtained using the trapezoidal (top row) and Simpson (bottom row) rules, see Eqs. (30) and (31) respectively.

(29) affects obtained numerical solutions. The order of accuracy of quadrature has visible but small impact on the obtained results. Differences between $\alpha_{T,dc}$ and $\alpha_{S,dc}$ obtained using Eqs. (30) and (31), respectively, are almost the same as differences between $\alpha_{T,d}$ and $\alpha_{S,d}$, compare results in Fig. 7.

The impact of $\epsilon_{h}(x,t)$ on $\alpha(\psi)$ can be summarized as follows. As it is expected, variation of $\epsilon_{h}(x,t)$ is affecting the shape of cumulative distribution function $\alpha(\psi)$. In the case of asymmetric, step shaped $\epsilon_{h,S}$ this finally leads to increment of the width of the intermittency region, see Fig. 4. One observes, the $\alpha(\psi)$ profile is approaching the equilibrium, analytical solution given by Eq. (13) with $\epsilon_h = \epsilon_{h,b}$, see Fig. 4 it = 45. We note, in the present numerical
Figure 7: The comparison of $\alpha(\psi)$ profile affected by the step (top row) or bell (bottom row) shaped variation of $\epsilon_h(x,t)$. Results are predicted using the trapezoidal ($\alpha_T$) or Simpson rules ($\alpha_S$) in the case of direct ($\alpha_{dc}$) or semi-analytical ($\alpha_d$) solutions at two selected time moments. The same results of $\alpha_{S,dc}$, $\alpha_{S,d}$ are depicted in Figs. 4 – 5.

Procedure the width of intermittency region is constrained by $\epsilon_{h,b}$, hence, $\epsilon_h$ variation can not result in the intermittency region thinner than $\epsilon_{h,b}$. In Fig. 4, the approximate solution $\alpha_d$ is reacting slightly faster on $\epsilon_{h,S}$ variation than $\alpha_{dc}$. However, $\alpha_d$ follows the direct solution $\alpha_{dc}$ very closely. The solutions $\alpha_{dc}$, $\alpha_d$ are bounded by the analytical $\alpha(\psi)$ profiles (black-dashed lines) with the extreme values of $\epsilon_h = \epsilon_{h,b}$ and $\epsilon_h = 2\epsilon_{h,b}$. The same conclusions can be drawn from the results presented in Fig. 5. During the non-symmetric changes of $\alpha(\psi)$ both, the exact and approximate solutions display similar behavior. Initially, resulting in the increase and then decrease of the intermittency region width. After the peak of $\epsilon_{h,B}(x,t)$ passes the expected position of the interface at $\psi=0$ the analytical $\alpha(\psi)$ profile with $\epsilon_h = \epsilon_{h,b}$ is recovered, see Fig. 5, $it = 45$. The
return of $\alpha (\psi)$ to the equilibrium state is guaranteed by the design of mapping function Eq. (27) and quadrature in Eq. (29).

The results presented in Figs. 4 – 5 confirm the semi-analytical solution $\alpha_d$ is providing close estimation of the coupled problem $\alpha_{dc}$. In the next section this solution is used to model variation of the intermittency region around circular drop without and with advection. As the third-order accurate Simpson quadrature is more sensitive to variations of $\epsilon_h (x, t)$ it is used in the all following numerical tests.

4.3. Two-dimensional semi-analytical solution

To assess how the numerical method introduced in Section 4.1 works in the two-dimensional case, the resting, circular drop centered at the point $(0.5, 0.5)$ with radius $R_B = 0.15$ [m] surrounded by the intermittency region and disturbed by the $\epsilon_{h,B} \leq \epsilon_h (x, t) \leq 5\epsilon_{h,B}$ field is studied, see Fig. 8. In this test, $\epsilon_h (x, t)$ is evolving according to Eq. (C.3). The problem is solved in a two-dimensional unit

![Figure 8: Evolution of the intermittency region affected by the variable, defined by Eq. (C.3) field of $\epsilon_{h,b} \leq \epsilon_h (x, t) \leq 5\epsilon_{h,b}$ (colors); all figures depict contours $\alpha(\psi = -4\epsilon_{h,b}), \alpha(\psi = 0), \alpha(\psi = 4\epsilon_{h,b})$.](image)
square box $[0, 1] \times [0, 1]$ discretized with $2^8 \times 2^8$ control volumes; the base width of the intermittency region is set to $\epsilon_{h,b} = \sqrt{2} \Delta x / 4 \,[m]$, the physical time step size is $\Delta t = 10^{-3} \,[s]$, fictitious time step size $\Delta \tau = \epsilon_{h,b} / 2 \,[s]$, four re-initialization steps $N_{r} = 4$ per $\Delta t$ are used. Only Eq. (18) with $\epsilon_{h,b} = \text{const.}$ and Eq. (28) taking into account $\epsilon_{h}(x, t)$ are solved as $w = u = 0$.

Figure 9: The comparison of the $\alpha(\psi)$ profiles affected by $\epsilon_{h}(x, t)$ field (see Fig. 8) with the analytical solutions. Diagrams present the $\epsilon_{h}(x, t)$ and $\alpha_{S,d}$ profiles drawn along the region $R1 \ a), b)$ and $R2 \ c), d)$ of the diagonal (see Fig. 8, it = 12) at seven time moments $it$. Black lines depict the analytical profiles of $\alpha(\psi)$ where $\epsilon_{h} = \epsilon_{h,b}$ (dashed line) and $\epsilon_{h} = 5 \epsilon_{h,b}$ (dashed-dotted line) obtained using Eq. (13).

The evolution of the intermittency region due to variable $\epsilon_{h}(x, t)$ is illustrated in Figs. 8 – 9. Fig. 8 displays variation of the $\epsilon_{h}(x, t)$ field and it impact on $\alpha(\psi)$ illustrated using contours $\alpha(\psi = -4 \epsilon_{h})$, $\alpha(\psi = 0)$, $\alpha(\psi = 4 \epsilon_{h})$. The variation of $\epsilon_{h}(x, t)$ in the subsequent time moments $it$ is leading first to increment, and afterwards to decrement of the intermittency region width to (similarly to
the one-dimensional predictions in Fig. 5). Details of this process can be observed in Fig. 9. Therein, $\epsilon_h(x,t)$ and $\alpha(\psi)$ profiles are drawn along the parts $R1, R2$ of the diagonal across the computational domain (see Fig. 8, $it=12$) at seven different time moments $it$. Fig. 9a,b is illustrating variations in the region $R1$ and Fig. 9c,d in the region $R2$.

In Fig. 9a, the impact of increasing in time $\epsilon_h(x,t)$ on $\alpha(\psi)$ is depicted. It can be observed, the profile of $\alpha(\psi)$ tends to its analytical solution given by Eq. (13) with $\epsilon_h=2.5\epsilon_{h,b}$ (not depicted in Fig. 9 for clarity of the presentation). After the bell shaped disturbance (moving to the left) passes $\psi=0$ (near $it=36$) the process is reversed and at $it = 60$ the equilibrium solution with $\epsilon_h = \epsilon_{h,b}$ is reconstructed, see Fig. 9b. Variation of $\epsilon_h(x,t)$ in the region $R2$ is more complex, see Fig. 9c,d. Therein, the step (moving to the left) and bell (moving to the right) shaped disturbances interfere, leading to increment of the $\alpha(\psi)$ profile width close to this obtained when $\epsilon_h = 5\epsilon_{h,b}$ in Eq. (13), see Fig. 9c, $it = 36$. In the subsequent time moments, the bell and step disturbances pass $\psi=0$ and the $\alpha(\psi)$ profile tends to the equilibrium solution where $\epsilon_{h,b}=2.5\epsilon_{h,b}$, see Fig. 9d $it > 36$.

One notes, during evolution in times $t, \tau$ the $\alpha(\psi)$ profile remains bounded between two extreme solutions obtained with $\epsilon_h = \epsilon_{h,b}$ (black dashed line) and $\epsilon_h = 5\epsilon_{h,b}$ (black dashed-dotted line), see Fig. 9. In the two- or three-dimensional cases the integration (29) is carried out along the normal coordinate $\psi$ in the local system attached to the each point of regularized interface $\gamma$. Thus, as in the one-dimensional case, reduction to the equilibrium solution in the points where $\epsilon_h = \text{const.}$ is guaranteed by the design of the quadrature (31).

4.4. Two-dimensional semi-analytical solution with advection

In this section, the semi-analytical approach described and verified in Sections 4.1 – 4.3 is used to reconstruct behavior of the intermittency region surrounding a two-dimensional circular drop with the radius $R = 0.15 \text{[m]}$ initially located at the point $(0.5, 0.35)$ and advected in the divergence-free, constant, circular velocity field $u = (u_1, u_2) = V_0/L(y-0.5, 0.5-x)$ where $V_0 = 1 \text{[m/s]}$ and
$L = 1 \, [m]$. The size of computational domain, number of control volumes and settings of the solver of Eqs. (17) and (18) are the same as described in Section 4.3. In order to obtain the full rotation of the drop in the given velocity field $u$, $\Delta t = 2.5 \cdot 10^{-3} \, [s]$ and $N_t = 2560$ time steps is required (the Courant number is $\sim 0.65$). The verification of the numerical methods and discretization of Eqs. (17) and (18) is described in details in the previous works of the present author (Waclawczyk, 2015, 2017). Herein, in Appendix D derivation of the Lagrangian scheme used to solve Eq. (17) in the known velocity field $u$ is recalled.

The intermittency region surrounding advected, circular interface is disturbed by the variable characteristic length scale field $\epsilon_h \,(x,t)$ defined using Eq. (C.6) as the linear superposition of Eqs. (C.1) and (C.2). The results depicted in Figs. 10 – 11 present subsequent time moments (from top to bottom) in the history of the advected circular bubble whose regularized interface is disturbed by variable $\epsilon_h \,(x,t)$ field. Each row in Figs. 10 – 11, shows the same three iso-contours $\alpha (\psi = -4\epsilon_{h,b})$, $\alpha (\psi = 0)$, $\alpha (\psi = 4\epsilon_{h,b})$ depicted against $\alpha (\psi)$, $\epsilon_h \,(x,t)$, $\psi (\alpha)$ fields, from left to right respectively. The third column in Figs. 10 – 11, presenting contours of the signed distance function $\psi (\alpha)$ is added to show the variation of cumulative distribution $\alpha (\psi)$ is predicted in the region where $n_\gamma$ is correctly defined, see Section 4.1 for description of the numerical method.

One observes, moving in the horizontal direction hat like profile has steep front and smooth tail, see Figs. 10 – 11. It interferes with the bell shaped axis-symmetrical characteristic length scale variation resulting in the increase of its local values, red color moving across the computational domain in the middle column of Figs. 10 – 11; the $\epsilon_h \,(x,t)$ field is bounded between $\epsilon_{h,b} < \epsilon_h \,(x,t) < 7\epsilon_{h,b}$. We note, introduced numerical model is sensitive to the rapid changes in the $\epsilon_h \,(x,t)$ field magnitude. For example, in Fig. 10, $it = 320$, 560 one observes (along the circumference of axis-symmetrical variation) how the width of the intermittency region is affected by variable $\epsilon_h \,(x,t)$. The proposed numerical method is sensitive to the local variation of $\epsilon_h \,(x,t)$ smoothness, too. In Fig. 11 $it = 1280$, one notes (along the left-right borders of the step disturbance) how
Figure 10: Evolution of the intermittency region affected by the variable field $\epsilon_h(x, t)$. All figures depict contours of $\alpha(\psi = -4\epsilon_{h,b})$, $\alpha(\psi = 0)$, $\alpha(\psi = 4\epsilon_{h,b})$, and from left to right fields of $0 < \alpha(\psi) < 1$, $\epsilon_{h,b} \leq \epsilon_h(x, t) < 7\epsilon_{h,b}$, contours $-32\epsilon_{h,b} \leq \psi(\alpha) \leq 32\epsilon_{h,b}$. 
Figure 11: Evolution of the intermittency region affected by the variable field $\epsilon_h(\mathbf{x},t)$.
All figures depict contours of $\alpha(\psi = -4\epsilon_{h,b})$, $\alpha(\psi = 0)$, $\alpha(\psi = 4\epsilon_{h,b})$, and from left to right fields of $0 < \alpha(\psi) < 1$, $\epsilon_{h,b} \leq \epsilon_h(\mathbf{x},t) < 7\epsilon_{h,b}$, contours $-32\epsilon_{h,b} \leq \psi(\alpha) \leq 32\epsilon_{h,b}$.
steepness of the $\epsilon_h(x,t)$ profile affects the $\alpha(\psi)$ field. Finally, as it can be observed in the last time moment presented in Fig. 11, in the domain where $\epsilon_h = \epsilon_{h,b}$ the intermittency region returns back to its original, equilibrium state. As it was mentioned previously this is achieved by the definition of quadrature (31) used to approximate Eq. (29).

5. Conclusions

In the present paper, the non-equilibrium model of the intermittency region between two weakly miscible phases is put forward. This new model is designed for the framework of one-fluid model of two-phase flow. At first, the evolution equation of the intermittency region is derived from the stochastic viewpoint, see Eq. (12). Next, based on the mesoscopic and macroscopic interpretations of the intermittency region the conditions of its equilibrium and non-equilibrium states are identified, see Section 2.1. The statistical interpretation of the solution to Eq. (12) is used to argue, the sharp interface tracked or captured in the VOF, SLS models is localized inside the mesoscopic intermittency region (gas-liquid macroscopic interface) remaining in the equilibrium state, see Section 2.2. It is explained, the level-sets $H_{\gamma} = 1/2, \psi = 0$ are two-dimensional representations of the expected position of the mesoscopic interface $\Gamma$ disturbed by thermal fluctuations. This result unfolds how the molecular effects are taken into account in the sharp models of the regularized interface $\gamma$ allowing them to predict the majority of topological changes initiated on the molecular level by stochastic, thermal fluctuations (Perumanath et al., 2019).

The equilibrium, sharp interface model is a good choice as long as the stochastic fluctuations of $\Gamma$ are small and/or do not depend on the background physical phenomena. In particular, in the case of turbulence/gas-liquid interface interactions the macroscopic intermittency region is expected to be in the non-equilibrium state due to spatiotemporal changes of turbulence statistics. For this reason, the second part of the present paper, is devoted to the case when the intermittency region may be in the non-equilibrium state as well. It
is demonstrated, finding the stationary solution to Eq. (12) when $\epsilon_h \neq const.$ is equivalent to minimization of the corresponding free energy functional, see Section 3.1. This result qualifies the stationary solution to Eq. (12) as the equilibrium condition accounting for the variable characteristic length scale $\epsilon_h(x,t)$. This equilibrium condition is used to derive the modified mapping between $\alpha(\psi) - \psi(\alpha)$ functions in Section 3.2. The new mapping function, allows to account for the non-equilibrium effects due to variable characteristic length scale $\epsilon_h(x,t)$ governing the intermittency region thickness and leads to the semi-analytical solution of the considered problem.

Finally, the numerical method permitting to account for the non-equilibrium effects is introduced in Section 4.1. First, the exact and semi-analytical solutions of Eq. (12) are compared and differences between them are discussed see Section 4.2. Afterwards, the semi-analytical solution of Eq. (12) is used to investigate the variations of $\alpha(\psi)$ due to $\epsilon_h(x,t)$ in tests without and with advection showing good agreement with the analytical solutions, see Fig. 9.

It is anticipated, the semi-analytical approach introduced in the present paper could be used with the existing numerical sharp/diffusive interface models to approximate the effects of intermittency region non-equilibrium on the flow field. The only requirement is reconstruction of the signed distance function field based on the known expected position of the gas-liquid interface. The modeling framework introduced herein, is planned to be used in future statistical models of the macroscopic interface agitated by turbulent fields, or mesoscopic interface affected by variable thermal energy, pressure and/or concentration variations.

Acknowledgments

This work is supported by the grant of National Science Center, Poland (Narodowe Centrum Nauki, Polska) in the project “Statistical modeling of turbulent two-fluid flows with interfaces”, ref. nr. 2016/21/B/ST8/01010, ID:334165.
Appendix A. Exact relations in the surface averaging

To derive Eq. (12) two exact relations between the ensemble $\langle \cdot \rangle$ and surface $\langle \cdot \rangle_{\Gamma \Sigma}$ averages are used. The first one is obtained directly from Eq. (3) as it implies

$$\langle W' \cdot \nabla H_{\Gamma} \rangle = \langle W' \cdot n_{\Gamma} \rangle_{\Gamma \Sigma}.$$  

(A.1)

The second exact relation $\langle n_{\Gamma} \rangle_{\Gamma \Sigma} = \nabla \langle H_{\Gamma} \rangle$ can be derived starting from the definition of the phase indicator function (Tryggvason et al., 2011)

$$H_{\Gamma} (x, y, z, t) = \int \int \int_V \delta (x - x') \delta (y - y') \delta (z - z') \, dx' dy' dz'$$  

(A.2)

and its gradient

$$\nabla H_{\Gamma} (x, y, z, t) = - \int \int_{\Gamma} (-n_{\Gamma}') \delta (x - x') \delta (y - y') \delta (z - z') \, d\Gamma'$$  

(A.3)

In Eq. (A.3) relation $\nabla \delta (x - x') = - \nabla' \delta (x - x')$ is used, moreover, it is noticed in the divergence theorem unit vector points outwards surface $\Gamma$ unlike the normal vector $n_{\Gamma} = \nabla \Psi / |\nabla \Psi|$. According to (Pope, 1998), Eq. (A.3) can be rewritten as

$$\nabla H_{\Gamma} (x, t) = \int \int_{\Gamma} n_{\Gamma} (\mu, \lambda, t) \delta (x - x'(\mu, \lambda, t)) A(\mu, \lambda, t) \, d\mu d\lambda$$  

(A.4)

where $\delta (x - x'(\mu, \lambda, t))$ is the three dimensional Dirac’s delta function. Therefore, in the local orthonormal coordinate system $\mu, \lambda, \Psi$ of infinitesimally small surface element $d\Gamma' = A(\mu, \lambda, t) \, d\mu d\lambda$, where $\Psi$ is the coordinate in the normal direction, Eq. (A.4) reads

$$\nabla H_{\Gamma} (\Psi) = \int \int_{\Gamma} n_{\Gamma} \delta (\mu) \delta (\lambda) \delta (\Psi - \Psi'(\mu, \lambda, t)) A(\mu, \lambda, t) \, d\mu d\lambda = \delta_{\Gamma}(\Psi) \, n_{\Gamma}$$  

(A.5)

additionally Eq. (A.5) allows to show

$$\frac{\partial H_{\Gamma} (\Psi)}{\partial \Psi} = \delta_{\Gamma}(\Psi).$$  

(A.6)

Using the surface average definition given by Eq. (4), the surface average of the normal vector $n_{\Gamma}$ is obtained as

$$\langle n_{\Gamma} \rangle_{\Gamma \Sigma} = \int \int_{\Gamma} \langle n_{\Gamma} \delta (\mu) \delta (\lambda) \delta (\Psi - \Psi'(\mu, \lambda, t)) A(\mu, \lambda, t) \rangle \, d\mu d\lambda$$  

(A.7)
where $\Sigma$ is defined by Eq. (5). Finally, taking the ensemble average $\langle \cdot \rangle$ of Eq. (A.5) and comparing with Eq. (A.7) results in the second exact relation
\[ \nabla \langle H_\Gamma \rangle = \langle n_\Gamma \rangle_\Gamma \Sigma. \] (A.8)

**Appendix B. Calculation of the free energy functional derivative**

In order to compute functional derivative of Eq. (19) with $\epsilon_h(x,t)$ we use the following definition. Given a functional $G [\alpha] = \int_V g(\psi,\alpha(\psi),\nabla \alpha(\psi)) \, dV$, (B.1) its functional derivative is obtained as
\[ \frac{\delta G}{\delta \alpha} = \frac{\partial g}{\partial \alpha} - \nabla \cdot \left[ \frac{\partial g}{\partial \nabla \alpha} \right]. \] (B.2)

The first term in Eq. (B.2), where $G = F$ and $F$ is given by Eq. (19), results in
\[ \frac{\partial f}{\partial \alpha} = \sigma \left[ \frac{2\alpha (1-\alpha)}{\epsilon_h} (1-2\alpha) + \frac{\delta k}{\delta \alpha} \right]. \] (B.3)

Since $\delta k/\delta \alpha$ is given by Eq. (20) and it does not depend explicitly on $\epsilon_h(x,t)$, Eq. (B.3) reads
\[ \frac{\partial f}{\partial \alpha} = 2\sigma \left[ \frac{(1-\alpha)(1-2\alpha)}{\epsilon_h} + \alpha \nabla \cdot n_\gamma \right]. \] (B.4)

The second term in Eq. (B.2), where $G = F$ and $F$ is given by Eq. (19), equals
\[ \nabla \cdot \left[ \frac{\partial f}{\partial \nabla \alpha} \right] = 2\sigma (\epsilon_h \nabla^2 \alpha + \nabla \epsilon_h \cdot \nabla \alpha). \] (B.5)

Therefore, to minimize the functional $F[\alpha]$, we search for
\[ \frac{\delta F}{\delta \alpha} = \epsilon_h \nabla^2 \alpha + \nabla \epsilon_h \cdot \nabla \alpha - \frac{\alpha (1-\alpha)}{\epsilon_h} [(1-2\alpha) + \epsilon_h \nabla \cdot n_\gamma] = 0. \] (B.6)

**Appendix C. Variations of the characteristic length scale field**

In the present work it is assumed variations of the characteristic length scale of the regularized interface $\epsilon_h(x,t)$ are known and given by the predefined formulas. The step profile
\[ \epsilon_{h,S}(x,t) = \epsilon_{h,b} \left[ 1 + \frac{H_S}{1 + \exp \left( - \frac{L(x,t)}{W_{\sigma \epsilon_h,b}} \right)} \right]. \] (C.1)
or the bell shaped profile

$$
\epsilon_{h,B}(x,t) = \epsilon_{h,b} \left[ 1 + H_B \exp \left( \frac{-f_B(x,t)}{W_B \epsilon_{h,b}} \right)^2 \right].
$$

(C.2)

In the one-dimensional cases presented in Figs. 4 – 5, \(H_S = H_B = W_S = 1, W_B = 6\) and \(\epsilon_{h,b} = \Delta x\). The functions \(f_S, f_B\) in Eqs. (C.1) and (C.2) are both set to \(f(x,t) = x - x_\Gamma + \epsilon_{h,b} (36 - it)\) where \(x_\Gamma = 0.5\) is position of the interface and \(it = 1, \ldots, 72\) denotes the physical time iteration number.

In the two dimensional case studied in Section 4.3, the variation of \(\epsilon_h(x,t)\) depicted in Figs. 8 – 9 is obtained as

$$
\epsilon_h(x,t) = \frac{1}{2} \left[ \epsilon_{h,S}(x,t) + \epsilon_{h,B}(x,t) \right]
$$

(C.3)

where in Eq. (C.2)

$$
f_B(x,t) = R - R_D + \epsilon_{h,b} (72 - it)
$$

(C.4)

and \(\epsilon_{h,b} = \sqrt{2} \Delta x/4, R = \sqrt{(x - r)^2}, r = (0.5, 0.5)\) determines the center and \(R_D = 0.2\) sets initial radius of axis-symmetrical part of \(\epsilon_h(x,t)\). In Eq. (C.3) the step, oblique variation is governed by

$$
f_S(x,t) = -(x-0.62) - (y - 0.62) - \epsilon_{h,b} \cdot it
$$

(C.5)

moreover \(H_B = H_S = 4, W_B = 8, W_S = 2, it = 1, \ldots, 16\).

In Section 4.4 where the semi-analytical solution is used together with the advection the following superposition of \(\epsilon_{h,B}\) and \(\epsilon_{h,S}\) is used

$$
\epsilon_h(x,t) = \frac{1}{2} \left[ \epsilon_{h,b} + \epsilon_{h,S1}(x,t) - \epsilon_{h,S2}(x,t) + \epsilon_{h,B}(x,t) \right]
$$

(C.6)

where \(\epsilon_{h,b} = \sqrt{2} \Delta x/4, \) variation of \(\epsilon_{h,B}\) with \(H_B = 5, W_B = 12\), is carried out using the function \(f_B(t)\) given by Eq. (C.4) where \(R_D = 0.2, r = (0.1, 0.1)\). Step profiles in Eq. (C.6) are defined using \(H_{S1} = H_{S2} = 3, W_{S1} = 1\) and \(W_{S2} = 10\), moreover, they are driven in time by the functions \(f_{S1}(x,t) = x - 0.15 - 0.25 \epsilon_{h,b} it\) and \(f_{S2}(x,t) = f_{1}(x,t) + 0.15\), respectively.
Appendix D. Derivation of Lagrangian scheme

The rearrangement of terms in the advection equation (17) leads to

$$\frac{1}{\alpha (1 - \alpha)} \frac{\partial \alpha}{\partial t} = -\frac{1}{\epsilon_h} |\nabla \psi| w \cdot n, \quad (D.1)$$

The left hand side is now integrated between $\alpha^n$ and $\alpha^{n+1}$, whereas the right hand side between $t^n$ and $t^{n+1}$ resulting in

$$\ln \left( \frac{\alpha}{1 - \alpha} \right) \bigg|_{\alpha^n}^{\alpha^{n+1}} = -\int_{t^n}^{t^{n+1}} \frac{1}{\epsilon_h} |\nabla \psi| w \cdot n \, dt, \quad (D.2)$$

where $n, n+1$ denotes old and new time levels, respectively. Integration given by Eq. (D.2) allows to derive the following scheme for advancement of $\alpha (\psi)$ in time $t$, given by the formula

$$\alpha^{n+1} = \frac{\alpha^n \exp [I(t^n)]}{1 - \alpha^n (1 - \exp [I(t^n)])}, \quad (D.3)$$

where the RHS integral in Eq. (D.2) is denoted as $I(t^n)$. This integral must be approximated by the appropriate quadrature; in the present work we adopt the second-order Adams-Bashfort method leading to

$$I(t^n) \approx -\left[ \frac{3}{2} f(t^n, \psi^n) - \frac{1}{2} f(t^{n-1}, \psi^{n-1}) \right] \Delta t, \quad (D.4)$$

where $f = |\nabla \psi| n_\gamma \cdot w / \epsilon_h$. The semi-analytical, explicit scheme given by Eqs. (D.3) and (D.4) is second-order accurate in time and no spatial discretization of $\alpha (\psi)$ is needed. It is noted, in the present work $\epsilon_{h+1} = \epsilon_h$ as the advection equation (17) is always solved with $\epsilon_{h, b} = \text{const.}$ in the semi-analytical case.

During numerical solution of Eq. (18) in time $\tau$, to obtain $\alpha_{dc}$ depicted in Figs. 4 – 5 and Fig. 7, the constrained interpolation (Waclawczyk, 2017) is used to determine $\tilde{\delta} (\alpha) = \alpha (1 - \alpha)$. The constrained interpolation in the present work is summarized below

$$\psi_f \approx \frac{1}{2} (\psi_P + \psi_F) + \mathcal{O} (\Delta x^2),$$

$$\alpha_f = \alpha (\psi_f) = \frac{1}{1 + \exp (-\psi_f I(\psi))}, \quad (D.5)$$

where subscripts $F, f, P$ denote the neighbor control volume $F$ and face $f$ of the given control volume $P$, respectively. $I(\psi)$ is the quadrature defined by Eq. (30) or Eq. (31) used to approximate integral (29).
Appendix E. Error norm

To show convergence during the numerical solution of Eq. (18) with $\epsilon_{h,B}(x,t)$, $\epsilon_{h,S}(x,t)$ (see Eqs. (C.1) and (C.2), respectively), in Fig. 6 for each physical time step $t$, $L_{1,\tau}$ error norm is plotted. This first-order norm is defined as

$$L_{1,\tau} = \frac{1}{N_c} \sum_{i=1}^{N_c} |\alpha_i^{n+1} - \alpha_i^n|,$$

(E.1)

where $N_c$ is the number of control volumes and $n+1$ denotes a new time level $\tau$, summation is performed over the control volumes centers in the entire computational domain.

References

Aarts, D. G. A. L., Schmidt, M., Lekkerkerker, H. N. W., 2004. Direct visual observation of thermal capillary waves. Science, 304, 847–850. doi:10.1126/science.1097116.

Allen, S., Cahn, J., 1979. A microscopic theory for antiphase domain boundary motion and its application to antiphase domain coarsening. Acta Metall., 27, 1085–1095.

Anderson, D. M., McFadden, G. B., Wheeler, A. A., 1998. Diffuse-Interface Methods in Fluid Mechanics. Annu. Rev. Fluid Mech., 30, 139–165. doi:10.1146/annurev.fluid.30.1.139.

Aniszewski, W., Boguslawski, A., Marek, M., Tyliszczak, A., 2012. A new approach to sub-grid surface tension for les of two-phase flows. Journal of Computational Physics, 231, 7368 – 7397. URL: http://www.sciencedirect.com/science/article/pii/S0021999112003890. doi:https://doi.org/10.1016/j.jcp.2012.07.016.

Brocchini, M., Peregrine, D. H., 2001a. The dynamics of strong turbulence at free surfaces. Part 1. Description. J. Fluid Mech., 449, 225–254.
Brocchini, M., Peregrine, D. H., 2001b. The dynamics of strong turbulence at free surfaces. Part 2. Free-surface boundary conditions. *J. Fluid Mech.*, *449*, 255–290.

Cahn, J. W., Hilliard, J. E., 1958. Free Energy of a Nonuniform System. I. Interfacial Free Energy. *J. Chem. Phys.*, *28*, 258–267. doi:https://dx.doi.org/10.1063/1.1744102.

Chiu, P.-H., Lin, Y.-T., 2011. A conservative phase field method for solving incompressible two-phase flows. *J. Comp. Phys.*, *230*, 185–204. doi:https://dx.doi.org/10.1016/j.jcp.2010.09.021.

Deike, L., Melville, W. K., Popinet, S., 2016. Air entrainment and bubble statistics in breaking waves. *Journal of Fluid Mechanics*, *801*, 91129. doi:10.1017/jfm.2016.372.

Elghobashi, S., 2019. Direct numerical simulation of turbulent flows laden with droplets or bubbles. *Annual Review of Fluid Mechanics*, *51*, 217–244. URL: https://doi.org/10.1146/annurev-fluid-010518-040401. doi:10.1146/annurev-fluid-010518-040401.arXiv:https://doi.org/10.1146/annurev-fluid-010518-040401.

Faust, J. A., 2018. Foreword. In J. A. Faust, and J. E. House (Eds.), *Physical Chemistry of Gas-Liquid Interfaces* Developments in Physical & Theoretical Chemistry (pp. Foreword, xvii). Elsevier. URL: http://www.sciencedirect.com/science/article/pii/B9780128136416120011. doi:https://doi.org/10.1016/B978-0-12-813641-6.12001-1.

Fedeli, L., 2017. Computer simulations of phase field drops on superhydrophobic surfaces. *J. Comp. Phys.*, *344*, 247–259. doi:https://dx.doi.org/10.1016/j.jcp.2017.04.068.

Gibbs, J. W., 1874. *On the equilibrium of heterogeneous substances*. Academy.

Gruszczyński, G., Mitchell, T., Leonardi, C., Laniewski-Wollek, L., Barber, T., 2020. A cascaded phase-field lattice boltzmann model for the simulation of incompressible, immiscible fluids with high density contrast.
Guo, X., Shen, L., 2010. Interaction of a deformable free surface with statistically steady homogeneous turbulence. *J. Fluid Mech.*, 658, 32–62.

Herrmann, M., 2013. A sub-grid surface dynamics model for sub-filter surface tension induced interface dynamics. *Computers & Fluids*, 87, 92–101. URL: http://www.sciencedirect.com/science/article/pii/S0045793013000637. doi:https://doi.org/10.1016/j.compfluid.2013.02.008. USNCCM Moving Boundaries.

Höhne, T., Vallée, C., 2009. Modelling of stratified two phase flows using an interfacial area density model. In *Computational Methods in Multiphase Flow V* (pp. 123–133). New Forest, United Kingdom.

Hong, W.-L., Walker, D., 2000. Reynolds-averaged equations for free surface flows with application to high-Froude-number jet spreading. *J. Fluid Mech.*, 417, 183–209.

Kajzer, A., Pozorski, J., 2020. A weakly compressible, diffuse-interface model for two-phase flows. *Flow, Turbulence and Combustion*, . URL: http://www.sciencedirect.com/science/article/pii/S0309561X19304732. doi:https://doi.org/10.1007/s10494-020-00164-8.

Komrakova, A. E., Eskin, D., Derksen, J. J., 2015. Numerical study of turbulent liquid-liquid dispersions. *AIChE Journal*, 61, 2618–2633. URL: https://aiche.onlinelibrary.wiley.com/doi/abs/10.1002/aic.14821. doi:10.1002/aic.14821. arXiv:https://aiche.onlinelibrary.wiley.com/doi/pdf/10.1002/aic.14821.

Labourasse, E., Lacanette, D., Toutant, A., Lubin, P., Vicent, S., Lebaigue, O., Caltagirone, J.-P., Sagaut, P., 2007. Towards large eddy simulation of
isothermal two-phase flows: Governing equations and a priori tests. *Int. J. Multiphase Flow*, 33, 1–39.

Lu, J., Tryggvason, G., 2018. Direct numerical simulations of multifluid flows in a vertical channel undergoing topology changes. *Phys. Rev. Fluids*, 3, 084401. URL: https://link.aps.org/doi/10.1103/PhysRevFluids.3.084401. doi:10.1103/PhysRevFluids.3.084401.

McCaslin, J. O., Desjardins, O., 2014. A localized re-initialization equation for the conservative level set method. *J. Comp. Phys.*, 262, 408 – 426. doi:http://dx.doi.org/10.1016/j.jcp.2014.01.017.

Olsson, E., Kreiss, G., 2005. A conservative level-set method for two phase flow. *J. Comp. Phys.*, 210, 225–246.

Osher, S., Fedkiw, R., 2003. *Level Set Methods and Dynamic Implicit Surfaces*. Springer Verlag, INC. New-York.

Osher, S., Sethian, J. A., 1988. Fronts propagating with curvature-dependent speed: Algorithms based on Hamilton-Jacobi formulations. *J. Comp. Phys.*, 79, 12 – 49. doi:http://dx.doi.org/10.1016/0021-9991(88)90002-2.

Perumanath, S., Borg, M. K., Chubynsky, M. V., Sprittles, J. E., Reese, J. M., 2019. Droplet coalescence is initiated by thermal motion. *Phys. Rev. Lett.*, 122, 104501. URL: https://link.aps.org/doi/10.1103/PhysRevLett.122.104501. doi:10.1103/PhysRevLett.122.104501.

Pope, S., 1998. The evolution of surfaces in turbulence. *Int. J. Eng. Sciences*, 26, 445–469.

Prosperetti, A., Tryggvason, G., 2007. *Computational Methods for Multiphase Flow*. Cambridge University Press.

Saeedipour, M., Schneiderbauer, S., 2019. A new approach to include surface tension in the subgrid eddy viscosity for the two-phase les. *International Journal of Multiphase Flow*, 121, 103128. URL: http:
Skartlien, R., Hu, B., Palmer, T., Staff, G., Sollum, E., 2014. A statistical model for the average volume fraction profile through the mixing zone in turbulent stratified gas-liquid flow. *International Journal of Multiphase Flow*, 59, 160 – 172. URL: [http://www.sciencedirect.com/science/article/pii/S0301932213001870](http://www.sciencedirect.com/science/article/pii/S0301932213001870). doi:10.1016/j.ijmultiphaseflow.2013.11.002.

Smolentsev, S., Miraghaie, R., 2005. Study of a free surface in open-channel water flows in the regime from ”weak” to ”strong” turbulence. *Int. J. Multiphase Flows*, 31, 921–939.

Soligo, G., Roccon, A., Soldati, A., 2019. Breakage, coalescence and size distribution of surfactant-laden droplets in turbulent flow. *Journal of Fluid Mechanics*, 881, 242–282. doi:10.1017/jfm.2019.772.

Sussman, M., Smith, K., Hussaini, M., Ohta, M., Zhi-Wei, R., 2007. A sharp interface method for incompressible two-phase flows. *Journal of Computational Physics*, 221, 469–505.

Thiesset, F., Duret, B., Mnard, T., Dumouchel, C., Reveillon, J., Demoulin, F. X., 2020. Liquid transport in scale space. *Journal of Fluid Mechanics*, 886, A4. doi:10.1017/jfm.2019.1056.

Toutant, A., Fournier, C., Chandesris, M., Jamet, D., Lebaigue, O., 2007. Interfacial conditions at a filtered interface for LES two-phase flows. In *Proceedings of the 6-th International Conference on Multiphase Flow*. Leipzig, Germany.

Tryggvason, G., Scardovelli, R., Zaleski, S., 2011. *Direct Numerical Simulations of Gas-Liquid Multiphase Flows*. Cambridge University Press.

Vrij, A., 1973. Light scattering from liquid interfaces. *Chemie Ingenieur Technik*, 45, 1113–1114. doi:10.1002/cite.330451807.
van der Waals, J., 1979. The thermodynamic theory of capillarity under the hypothesis of a continuous variation of density. *J. Statist. Phys.*, 20, 200–244.

Waclawczyk, M., Oberlack, M., 2011. Closure proposals for the tracking of turbulence-agitated gas-liquid interfaces in stratified flows. *Int. J. Multiphase Flow*, 37, 967–976.

Waclawczyk, M., Waclawczyk, T., 2015. A priori study for the modelling of velocity-interface correlations in the stratified air-water flows. *Int. J. Heat Fluid Flow*, 52, 40 – 49. doi:http://dx.doi.org/10.1016/j.ijheatfluidflow.2014.11.004.

Waclawczyk, T., 2015. A consistent solution of the re-initialization equation in the conservative level-set method. *J. Comp. Phys.*, 299, 487 – 525. doi:http://dx.doi.org/10.1016/j.jcp.2015.06.029.

Waclawczyk, T., 2017. On a relation between the volume of fluid, level-set and phase field interface models. *International Journal of Multiphase Flow*, 97, 60 – 77. URL: http://www.sciencedirect.com/science/article/pii/S0301932216307856. doi:https://doi.org/10.1016/j.ijmultiphaseflow.2017.08.003.

Waclawczyk, T., Waclawczyk, M., Kraheberger, S. V., 2014. Modeling of turbulence-interface interactions in stratified two-phase flows. *Journal of Physics: Conference Series*, 530.