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Modeling dispersed phase holdup and conditions of flooding in a pulsed disc and doughnut solvent extraction column

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Abstract

Pulsed disc and doughnut solvent extraction columns (PDDCs) have emerged as an efficient alternative to conventional mixer-settlers and pulsed sieve plate extraction columns for nuclear spent fuel extraction. This study discusses the feasibility of the turbulent volume of fluid (VOF) and hybrid multi-fluid Eulerian multiphase methods to predict the dispersed phase holdup and flooding conditions of an experimental PDDC with 7.5-cm internal diameter. Analysis of axial holdup and axial velocity was incorporated to rationalize the droplet breakage and coalescence along the column. The onset of local and total flooding was modeled and compared using both methods. The evolution of recirculating zones revealed insightful information about local holdup distributions during flooding. Unlike conventional Eulerian multiphase modeling, the VOF method described accurately the holdup and the onset of local flooding, but a finer mesh was required. Alternatively, the hybrid multi-fluid Eulerian method could capture accurately turbulent length scales while predicting the holdup and flooding conditions, but required optimization of the dispersed phase droplet diameter through multiple iterations.

Keywords: Pulsed disc and doughnut extraction column; Volume of fluid method; Hybrid multi-fluid Eulerian approach; Dispersed phase holdup; Flooding; Multiphase simulation.

Glossary

Variables

a_1 :	Model constant
A :	Pulsing amplitude (m)
AARE:	Average absolute relative error (%)
C_D :	Drag coefficient
C_α :	Artificial compression factor
$C_{Dk\omega}$:	Cross diffusion term
C_{vm} :	Virtual mass force coefficient
$C_{1\epsilon}, C_{2\epsilon}, C_\mu$:	Empirical model constant
d_a :	Doughnut aperture (m)
d_{32} :	Sauter mean diameter (m)
d :	Droplet diameter (m)
f :	Pulsing frequency (s^{-1})
f :	Volumetric surface tension force ($kg.m^{-2}.s^{-2}$)
f_b :	Body force per unit mass ($m.s^{-2}$)
$F_{D,k}$:	Drag force for phase k ($kg.m.s^{-2}$)
$F_{S,k}$:	Surface tension force for phase k ($kg.m.s^{-2}$)
$F_{vm,k}$:	Virtual mass force for phase k ($kg.m.s^{-2}$)
F_1 :	First blending function
F_2 :	Second blending function
$\mathbf{g}$:	Gravitational vector ($m.s^{-2}$)
G :	Turbulent kinetic energy generation ($kg.m^2.s^{-2}$)
h_c :	Compartment height (m)
k :	Turbulent kinetic energy ($kg.m^2.s^{-2}$)
L :	Length scale (m)
$\hat{\mathbf{n}}$:	Unit normal vector (-)
p :	Pressure (Pa)
R :	Organic-to-aqueous phase volume ratio
Re :	Reynolds number
S :	Shear strain
$\mathbf{S}_f$:	Area vector of a particular phase (m^2)
$\mathbf{U}$:	Velocity vector ($m.s^{-1}$)
V_c :	Continuous phase superficial velocity ($m.s^{-1}$)
V_d :	Dispersed phase superficial velocity ($m.s^{-1}$)
t :	Time (s)

x:	Interface position (m)
y:	Shortest distance to the next surface (m)

Greek symbols

α :	Volume fraction (-)
α, β, β^* :	Empirical model constants
ε :	Turbulent energy dissipation rate ($\text{m}^2 \cdot \text{s}^{-3}$)
κ :	Surface curvature
μ :	Viscosity ($\text{Pa} \cdot \text{s}^{-1}$)
ρ :	Liquid density ($\text{kg} \cdot \text{m}^{-3}$)
σ :	Surface tension ($\text{N} \cdot \text{m}^{-1}$)
$\sigma_k, \sigma_\epsilon, \sigma_\omega$:	Turbulent Prandtl numbers (-)
τ :	Shear stress ($\text{N} \cdot \text{m}^{-2}$)
ω :	Specific dissipation rate of omega (s^{-1})
ψ :	Specific power consumption ($\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-3}$)

Subscripts

cont:	Continuous phase
dis:	Dispersed phase
eff	Effective
r:	Relative
T:	Turbulent

Superscripts

*:	Modified
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Acronyms

CFD:	Computational Fluid Dynamics
d.c:	Duty cycle
HTU:	Height of transfer units
RDC:	Rotating disc contactor
PDDC:	Pulsed disc and doughnut extraction column
PSPC:	Pulsed sieve plate column
STT:	Shear Stress Transport
TBP:	Tri butyl phosphate
VOF:	Volume of Fluid

1. Introduction

Pulsed extraction columns are a major work force in the solvent extraction industry [1]. They are mostly applied in nuclear fuel recycling, hydrometallurgical operations [2,3] and for metal recycling in batteries [4], and can be equally efficient in food processing, cosmetics and medical fields. Opposing mixer-settlers and other well-established multistage extraction devices like Kuhni columns and rotating disc contactors (RDC), pulsed extraction columns exhibit much safer operation since the imparted energy can be controlled remotely and without complex machinery involving moving parts like impellers [5]. The to and fro pulsing action inherent to pulsed extraction columns makes them more efficient than conventional packed columns by reducing the height of transfer units (HTUs) from three to five times [6]. Moreover, unlike mixer settlers, they require much less foot-space and organic loading that prevents unwanted solvent degradation and loss.

Pulsed extraction columns can be broadly classified in two families, namely pulsed sieve plate columns (PSPC) and pulsed disc and doughnut extraction columns (PDDC). Both column designs are used in nuclear and hydrometallurgical applications. PDDCs are more adapted to feeds with higher amount of dissolved solids due to their geometrical configuration and display lower mass transfer resistances than PSPCs due to their higher interfacial areas [7]. This study focuses on PDDCs and targets the development of robust and advanced computational methods to simulate and predict their performance and hydrodynamic regimes, namely the mixer-settler, dispersion and quasi-emulsion regimes [8–11]. The mixer-settler regime occurs at low pulsation intensity where the secondary (organic) phase initially breaks into larger organic droplets with the upward stroke of pulsing, and then settles to form a layer beneath the discs with the downward stroke of the pulsing cycle. As the pulsation intensity increases, the settled layers breaks into larger droplets which have a higher settling velocity and as a result the holdup decreases [8]. This phenomenon changes the column's hydrodynamic regime from the mixer-settler to the transition regime. Mass transfer is poor in both regimes due to insufficient interfacial area and, accordingly,

they are not preferred for operating PDDCs [12,13]. By increasing the pulsation intensity, the droplets become smaller and well dispersed, and the column enters the dispersion regime. In this regime, due to the reduction of droplet sizes, the holdup increases encompassing a much promoted mass transfer [14]. Further increase of the pulsation intensity results in a sustained decline of the droplet size, and the column enters the quasi-emulsion regime where mass transfer is marginally reduced due to the formation of finer droplets although higher bubble throughputs can be maintained.

As a rule, the performance of PDDCs is described using empirical correlations where key hydrodynamic parameters such as the dispersed phase holdup, Sauter mean diameter, slip velocity and flooding / characteristic velocities are described as a function of the operation variables, physical properties of liquid phases, column geometry and wettability of plates [5,8,11,14–24]. These correlations rely on many parameters that makes understanding of the role of operation variables in the column performance and upscaling not straightforward. In this context, numerical investigations are necessary to understand the flow fields and energy distributions in the columns, and above all the intricate physics of multiphase flow [25].

Very few numerical studies have been reported to date focusing on PDDCs including single phase and multiphase flows. Starting from the analysis of unsteady laminar single-phase flow predictions [26], modeling steady-state turbulent flow was first attempted using an Eulerian model with a standard k-epsilon turbulence model [27–29].

Subsequently, axial dispersion in steady [30] and turbulent conditions [31–34] was incorporated leading to accurate estimates of experimental axial dispersion coefficients in a PDDC. Calculations of discrete particle residence time distributions with single-phase hydrodynamics based on a Lagrangian formulation have also been reported [35,36]. However, this approach incorporates only momentum transfer from fluid to particles (i.e. one-way Lagrangian coupling) and proves inadequate to reproduce highly dispersed phase holdup flows (>10%) commonly found in PDDC industrial columns [37]. In this view, it is imperative to assess

the impact of the dispersed phase on the continuous phase (two-way Lagrangian coupling). Overall, calculations based on Lagrangian models require to fully resolve droplet-droplet and droplet-wall interactions, and as such are very expensive and difficult to implement to model the hydrodynamics of PDDCs.

To overcome the limitations of Lagrangian formulations, two-phase Eulerian models that treat each phase as an interpenetrating continuum have become popular to model the dispersed phase holdup in PDDCs [33,38–41]. Recently, the traditional Eulerian multiphase approach has evolved by incorporating droplet breakage and coalescence using population balance modeling and Monte Carlo simulations to predict the mean droplet diameter [42–44]. However, in these studies, the surface tension force was not included in the momentum equation which is a key parameter governing the performance of liquid-liquid extraction systems. Very recently, the Eulerian approach was modified for liquid-liquid systems with sharp interfaces by adding an extra artificial compression term in the phase fraction transport equation [45]. Overall, although the above approaches are relatively inexpensive compared to two-way coupled, fully resolved Lagrangian simulations, they remain expensive since the continuity and momentum equations need to be solved simultaneously for each phase along with the interphase exchange terms including drag, lift and virtual mass forces.

To overcome the intrinsic limitations of multiphase models based on Eulerian formulations to describe the hydrodynamics of PDDCs, the volume of fluid (VOF) method is a powerful approach where the two-fluid system is treated as ‘one effective fluid’ in an Eulerian formulation and thus only one continuity and momentum equation along with turbulence models are required to describe the liquid-liquid system [46,47]. This allows a significant reduction of the computational cost. As a rule, simulations by the VOF method are about five times less expensive than conventional Eulerian methods and differ from them only marginally in terms of mean velocity and volume fraction predictions [48]. These differences stay well below experimental errors.

Despite its promising credentials, the VOF method has been seldom applied to simulate bubble column dynamics [49–52] and, to our knowledge, no report is available describing the hydrodynamics and mass transfer in liquid-liquid extraction devices. This paper aims to fill this gap and provide a critical appraisal of the VOF method to predict the overall and axial dispersed phase holdup, flooding conditions, and turbulence parameters like the turbulent kinetic energy and its dissipation rate of PDDCs. For comparison, we conducted calculations using the hybrid Eulerian multiphase model developed by Wardle *et al* [45] as it incorporates interface tracking to capture sharp interfaces between the continuous and dispersed phases together with surface tension forces, and is less computationally expensive than more conventional population balance Eulerian approaches. The validity of the two methods was assessed by comparison of predicted trends with experimental holdup and flooding data measured in a laboratory scale PDDC (7.5-cm diameter; 2-m active section height; 70 pairs of disc and doughnut internals with 23% free area) for the extraction 3 M nitric acid solution in 30% tributyl phosphate (TBP) with dodecane. The experiments were carried out at four levels of total superficial velocity and pulsation intensities covering the mixer-settler and quasi-emulsion regimes that are commonly found in PDDCs for nuclear fuel reprocessing.

2. Materials and Methods

2.1. Materials

A 3-M aqueous nitric acid solution (aqueous phase) was used as continuous phase (density: 1100 kg/m³; viscosity: 1.001 mPa·s). TBP (density: 980 kg/m³; 99% pure) and n-dodecane (density: 760 kg/m³; 99% pure) were used to prepare the disperse phase (30%v/v TBP in n-dodecane; density: 838 kg/m³; viscosity: 1.930 mPa·s; interfacial tension: 10.38 mN/m). All the materials were supplied by the plutonium plant at the Bhabha Atomic Research Centre, Trombay. Distilled water (0.5-3 µS/cm) was used to prepare nitric acid solutions at different concentrations. A Du Nouy ring tensiometer (TD2- Lauda) was used to estimate the interfacial tension. Density was calculated by a hydrometer and viscosity was measured by SVM-3000 Stabinger viscometer.

2.2. Experimental setup

The experimental setup and the scheme of the PDDC implemented in this study are depicted in **Figure 1**. Additional details on the column design and flowsheet are provided in the SI (**Figure S1**) and in a previous report [53]. Briefly, the column consisted of a glass column (diameter: 7.5 cm; extraction section height: 2 m) with two end settlers (diameter: 18.14 cm; height: 45 cm) and 70 pairs of disc and doughnut internals in alternate patterns resulting in a serpentine flow through the doughnuts and annular sections beside the discs. The internals were built in stainless steel (SS304L) and had a thickness of 2 mm. The reason for choosing SS304L was due to its preferential wettability towards the aqueous phase (contact angle close to 90 degrees at room temperature [54]). This property of SS304L facilitated droplet formation for the dispersed organic phase). The inter-plate spacing was set to 12.5 cm and the plates were held together with three tie rods and spacers (diameter: 3.2 mm). The column had two inlet and two outlet valves for the aqueous and organic phases. The pulsing energy was imparted through a pulse leg (diameter: 5.0 cm). A double acting three-way solenoid valve (diameter: 5.0 cm) was used to facilitate the pulsing action inside the column using compressed air. To mitigate the compressor disturbance a capacitance air reservoir (20 L) was provided just before the solenoid.

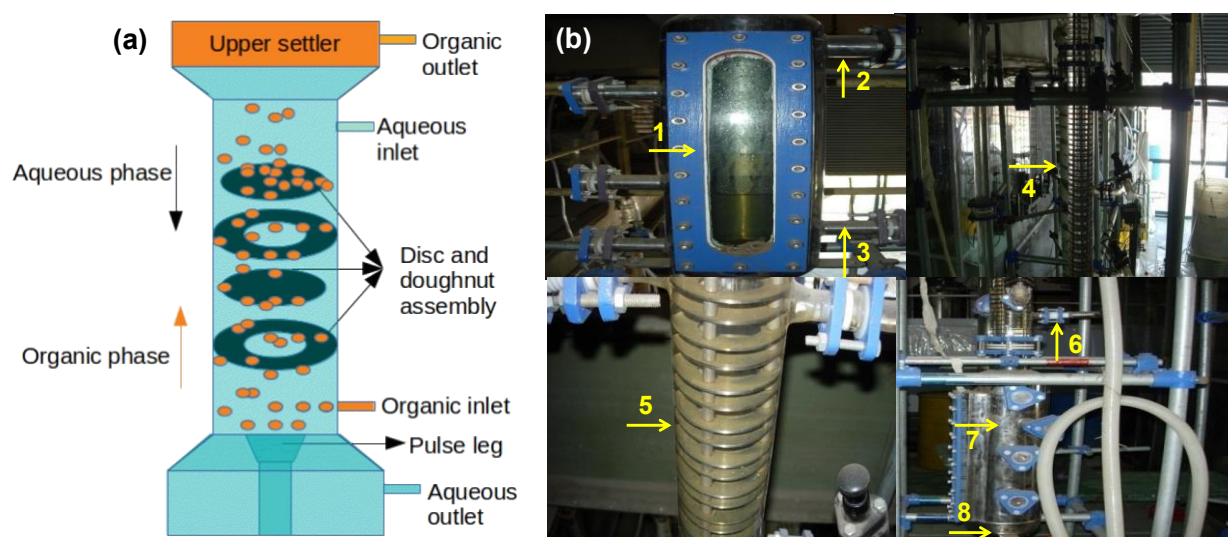


Figure 1. (a) General scheme of the PDDC implemented in this study. (b) Different parts in the PDDC used in this study: (1) upper end settler, (2) organic outlet, (3) aqueous inlet, (4) active

section of the column, (5) disc and doughnut assembly with tie rods, (6) organic inlet, (7) lower end settler and (8) aqueous outlet.

2.3. Methodologies

2.3.1. Measurement of dispersed phase holdup

The dispersed phase holdup was measured by the volume displacement method according to a previous report [14]. In these tests, the column was filled first with the aqueous phase and the pulsing amplitude, frequency and percentage duty cycle were set to define the desired pulsing velocity profile in the longitudinal direction (**Figure 2**). Each pulsating cycle (covering 1 s) was divided in two sections that were denoted as *upward* and *downward* referring to the time duration when the solenoid pushed air inside the column through the pulse leg or released air, respectively. From both sections, the percentage duty cycle was defined as the time percentage of the upward section in a pulsating cycle. For example, a duty cycle of 0.5 means 50% of the pulsating cycle time comprised the upward section while the remaining time comprised the downward section. Therefore, the time duration of the upward stroke of solenoid decreased at lower percentage duty cycles and, to keep the same pulsing amplitude, a larger reservoir pressure was required resulting in an increased turbulence in the column.

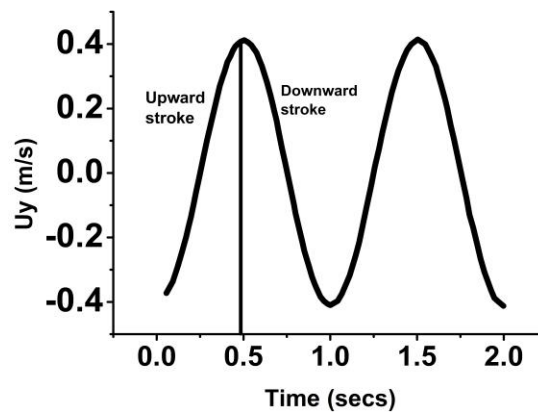


Figure 2. Pulsing velocity profile in the longitudinal direction in the PDDC.

After setting the pulsating cycles, the secondary (organic) phase was introduced keeping the pulsing on until the interface at the upper end settler attained steady state. At this point, all the

inlet and outlet valves were closed, and the pumps were shut down. The pulsing was continued until all the inter-plate organic phases reached the liquid-liquid interface in the upper end settler. The change in the interface height was then measured and the organic phase volume was calculated which was then divided by the volume of the extraction section to obtain the fractional dispersed phase holdup. The experimental error in the measured dispersed phase holdup was $\pm 5\%$.

2.3.2. Measurement of flooding conditions

To measure the throughput at the onset of flooding and at total flooding, the total superficial velocity was increased by keeping the pulsation intensity constant. The onset of flooding was characterized by the formation of globules of organic dispersed phase combined with a rapid increase in the dispersed phase holdup. The dispersed phase holdup was steady until the condition of total flooding was achieved due to local layer formation of the dispersed phase. At this stage, the holdup was observed to shoot up significantly and the column became inoperable. The dispersed phase holdup was measured only at the onset of flooding.

2.3.3. Operation variables

The dispersed phase holdup was measured at continuous (V_c) and dispersed phase superficial velocities (V_d) in the range 0.125-0.375 cm/s at different pulsation intensities ($A*f$; A -pulsing amplitude; f - pulsing frequency) in the range 0.936-1.872 cm/s keeping the organic-to-aqueous phase volume ratio at 1. This range of superficial velocities covered the mixer-settler regime (V_c and V_d : 0.125-0.25 cm/s; Af -0.936-1.25 cm/s) to quasi-emulsion regime (V_c and V_d : 0.314-0.375 cm/s; Af -0.936-1.872 cm/s). The percentage duty cycle (d.c) was kept at 35%. To assess the flooding conditions, V_c and V_d was varied at a pulsation intensity of 1.56 cm/s.

3. Computational methodologies

Computational modeling of the dispersed phase holdup and flooding conditions in PDDCs was carried out using the VOF method. To assess its robustness, we also modeled PDDCs using the

hybrid multiphase Eulerian method. Unlike conventional Eulerian methods, this latter method incorporates interface tracking and surface tension forces. Turbulence was implemented in both methods using the k-epsilon and k-omega-SST (SST = Shear Stress Transport) models. Both methods are briefly discussed in section 3.1 along with the turbulence models.

3.1. Models and governing equations

3.1.1. Representation of the liquid-liquid interface

In liquid-liquid extraction, the size and stability of dispersed phase droplets depend mainly on the density and viscosity of the aqueous and organic phases along with the interfacial tension. In the VOF method, the volume fraction of the dispersed phase, α , was tracked in each cell to ascertain the position and attributes of the interface [55]. **Figure 3** plots a typical representation of an interface between the continuous and dispersed phases, where the colored and uncolored areas represent the dispersed and continuous phases, respectively. The numerical value in each cells denotes the value of α that is comprised between 0 and 1:

$$\alpha(x, t) = \begin{cases} 0, & \text{continuous phase} \\ 0 < \alpha < 1, & \text{interface} \\ 1, & \text{dispersed phase} \end{cases} \quad (1)$$

where, x is the position of the interface at time t .

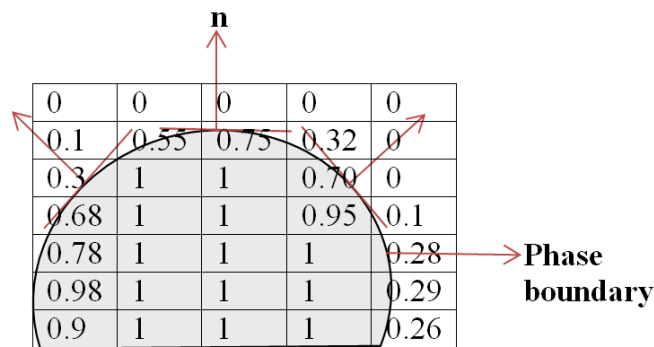


Figure 3. Pictorial representation of an interface in the VOF method.

In both methods, the liquid phases were regarded as a single effective liquid and only one continuity and one momentum equation were solved. The surface tension force was included in the momentum equation as a source term to capture the liquid-liquid interface by applying the

continuum surface model [56]. The density and viscosity of the effective liquid were calculated by adding the continuous and dispersed phase densities and viscosities weighted by their respective phase fractions. To reconstruct the interface, a scalar transport equation of the volume fraction was solved, and the relative velocity between the phases was implemented to retain the two liquid natures (see details in section 3.1.2). This relative velocity was calculated by introducing an artificial compression factor that acted only at the proximity of the liquid-liquid interface ($0 < \alpha < 1$) to reduce the interfacial instability and the effect of numerical oscillations [57]. The advection equation was solved using the flux corrected technique allowing a precise interfacial modeling [47].

3.1.2. VOF method

The continuity and momentum equations for incompressible and immiscible phases were simultaneously solved together with a scalar transport equation for α as given in Eq 2-4

$$\nabla \cdot \mathbf{U} = 0 \quad (2)$$

$$\frac{\partial \rho \mathbf{U}}{\partial t} + \nabla \cdot (\rho \mathbf{U} \mathbf{U}) = -\nabla p^* + \nabla \cdot (\mu_{\text{eff}} \nabla \mathbf{U}) + \mathbf{f}_b \quad (3)$$

$$\frac{\partial \alpha_{\text{cont.}}}{\partial t} + \nabla \cdot (\alpha_{\text{cont.}} \mathbf{U}) = 0 \quad (4)$$

where $\mathbf{U}$ is the velocity vector, ρ is the effective liquid density, μ_{eff} is the effective turbulent viscosity, p is the pressure and the *cont* suffix refers to the continuous phase. The velocity vector $\mathbf{U}$ is the shared velocity between phases and was expressed by Eq 5

$$\mathbf{U} = \mathbf{U}_{\text{cont.}} = \mathbf{U}_{\text{disp}} \quad (5)$$

The effective turbulent viscosity comprises the turbulent viscosity (μ_T) and the bulk fluid viscosity (μ) and was expressed by Eq 6

$$\mu_{\text{eff}} = \mu_T + \mu \quad (6)$$

In Eq 3, $\mathbf{f}_b$ is the body force per unit mass which includes gravity and surface tension forces, and was expressed by Eq 7

$$\mathbf{f}_b = \mathbf{g} \cdot \mathbf{x} \nabla \rho + \mathbf{f} \quad (7)$$

where $\mathbf{g}$ is the gravitational constant and $\mathbf{f}$ is the volumetric surface tension force.

The effective properties (density and viscosity) of the *one effective liquid* in the domain was expressed as a function of weighted averages based on the volume fraction of the continuous phase (Eq 8-9)

$$\rho = \rho_{\text{cont.}} * \alpha_{\text{cont.}} + \rho_{\text{disp.}} * (1 - \alpha_{\text{cont.}}) \quad (8)$$

$$\mu = \mu_{\text{cont.}} * \alpha_{\text{cont.}} + \mu_{\text{disp.}} * (1 - \alpha_{\text{cont.}}) \quad (9)$$

where *disp* suffix denotes the dispersed phase.

Regarding Eq 9, a volume fraction weighted average method was used to calculate the mixture viscosity. Although Rajendran *et al* [58] argued that Eq 9 might be questionable for systems with large relative viscosity, this equation is still applicable for systems with small relative viscosities. In our case, the difference between the continuous and dispersed phase viscosities was only 0.92 mPa-s which is small. Moreover, reported studies related to the rising of single droplets in a quiescent liquid recommend the use of Eq 9 to calculate the viscosity [59]. Therefore, we used Eq 9 to calculate the mixture viscosity.

In the VOF method, accurate calculation of the phase fraction is of utmost importance to properly describe the surface curvature that affects the surface tension force and pressure gradient across the free surface. Although it is not trivial to ensure boundedness and conservativeness of the phase fraction, several attempts have been made to tackle the issue [60–63]. Here, the interface was tracked by solving a modified scalar transport equation of α according to Eq 10 (instead of Eq 4)

$$\frac{\partial \alpha_{\text{cont}}}{\partial t} + \nabla \cdot (\alpha_{\text{cont}} \mathbf{U}) + \nabla \cdot (\alpha_{\text{cont}} * (1 - \alpha_{\text{cont}}) * \mathbf{U}_r) = 0 \quad (10)$$

where $\mathbf{U}_r$ is the relative velocity between the liquid phases and was calculated by Eq 11

$$\mathbf{U}_r = C_\alpha * |\mathbf{U}| * \frac{\nabla \alpha_{\text{cont}}}{|\nabla \alpha_{\text{cont}}|} \quad (11)$$

where, C_α is the artificial compression term supplied by the user, and $\frac{\nabla \alpha_{\text{cont}}}{|\nabla \alpha_{\text{cont}}|}$ estimates the unit normal vector to the interface for the cells where the condition $0 < \alpha_{\text{cont}} < 1$ is satisfied.

The volumetric surface tension force was computed from the continuum surface force model [56] as given in Eq 12

$$\mathbf{f} = \sigma * \boldsymbol{\kappa} * \nabla \alpha_{\text{cont}} \quad (12)$$

where σ is the surface tension and $\boldsymbol{\kappa}$ is the surface curvature that was calculated by Eq 13

$$\boldsymbol{\kappa} = \frac{\nabla \alpha_{\text{cont}}}{|\nabla \alpha_{\text{cont}}|} \quad (13)$$

In this method, the individual droplets rise because of the buoyancy force due to density difference between the continuous and dispersed phase. During the rise, the droplets are subjected to buoyancy, viscous and surface tension forces which cause the change in their shape from spherical (in 3D) and circular (in 2D) to oblate (in 3D) and elliptical (in 2D) depending on the diameter [59] and the hydrostatic head. Finally, after reaching 1-2 control volume to the liquid-liquid interface, the droplet coalesced.

3.1.3. Hybrid Eulerian multiphase method

In the hybrid Eulerian multiphase method, interface capture is incorporated to resolve droplet dispersions with sharp interfaces. Opposing traditional Eulerian multiphase models, this method allows a more realistic description of droplet dispersion in a continuous liquid phase. It is due to the fact that interface capture is applied where grid resolution is sufficient to resolve larger droplets with negligible inter-dispersion, and dispersed flow models are used where the droplets enter in the sub-grid scale. Cerne *et al* [64] devised a method for switching between the VOF and Eulerian models based on the gradient of volume fraction across the neighboring cells by solving the required number of model equations, but it resulted in a complex numerical procedure. To overcome this shortcoming, the authors opted for multi-fluid VOF coupling by adding an interface-capturing equation with mass conservation equation, and thereby the Eulerian

multiphase equations were solved in the entire domain and interface sharpening method was applied only at sharp interface regions. Later, Wardle *et al* [45] extended the approach to multistage solvent extraction devices such as centrifugal extractors showing the presence of large, dispersed phase droplets.

The mass and momentum equations for each phase i for the multi-fluid hybrid Eulerian method were expressed by Eq 14-15

$$\frac{\partial \alpha_i}{\partial t} + \mathbf{U} \cdot \nabla \alpha_i + \nabla \cdot (\alpha_i * (1 - \alpha_i) * \mathbf{U}_r) = 0 \quad (14)$$

$$\frac{\partial \rho_i \alpha_i \mathbf{U}_i}{\partial t} + (\rho_i \alpha_i \mathbf{U}_i \cdot \nabla) \mathbf{U}_{ik} = -\alpha_i \nabla p^* + \nabla \cdot (\mu_{i,\text{eff}} \alpha_i \nabla \mathbf{U}_i) + \rho_i \alpha_i \mathbf{g} + \mathbf{F}_{D,i} + \mathbf{F}_{S,i} + \mathbf{F}_{vm,i} \quad (15)$$

where α_i , $\mathbf{U}_k$ and ρ_i are the phase fraction, velocity and density of phase k , respectively, $\mathbf{g}$ is the gravity vector, and $\mathbf{F}_{D,i}$, $\mathbf{F}_{S,i}$ and $\mathbf{F}_{vm,i}$ are the drag, surface tension and virtual mass forces for phase k , respectively, defining the interphase momentum transfer force. In Eq 14, the term $[\nabla \cdot (\alpha_i * (1 - \alpha_i) * \mathbf{U}_r)]$ was added to capture the interface where $\mathbf{U}_r$ is the relative velocity between phases and calculated by Eq 11.

The drag force coefficient was formulated by Eq 16 according to Schiller and Naumann [65]

$$C_D = \begin{cases} \frac{24(1+0.15\text{Re}^{0.683})}{\text{Re}}, & \text{Re} \leq 1000 \\ 0.44 & \text{Re} > 1000 \end{cases} \quad (16)$$

where Re is the Reynold's number and is given by Eq 17

$$\text{Re} = \frac{|(\mathbf{U}_{\text{cont}} - \mathbf{U}_{\text{disp}})| * d_{\text{disp}}}{\nu_{\text{cont}}} \quad (17)$$

In parallel, the drag force equations were given by Eq 18-19

$$\mathbf{F}_{D,k} = \frac{3}{4} * \rho_{\text{cont}} * \alpha_{\text{cont}} * \alpha_{\text{disp}} * C_D * \frac{|(\mathbf{U}_{\text{cont}} - \mathbf{U}_{\text{disp}})| * (\mathbf{U}_{\text{cont}} - \mathbf{U}_{\text{disp}})}{d_{\text{disp}}} \quad (18)$$

$$\mathbf{F}_{D,k} = K * \alpha_{\text{cont}} * \alpha_{\text{disp}} * (\mathbf{U}_{\text{cont}} - \mathbf{U}_{\text{disp}}) \quad (19)$$

$$\text{where } K = \frac{3}{4} * \rho_{\text{cont}} * C_D * \frac{|(\mathbf{U}_{\text{cont}} - \mathbf{U}_{\text{disp}})|}{d_{\text{disp}}} \quad (20)$$

The dispersed phase Sauter mean drop diameter ranged from 1.85 to 4 mm resulting in a droplet Reynold's number in the range 4.6-26 (Eq 17) that was well below 1000 (Eq 16). The drag coefficient was estimated for each phase by considering each one as dispersed, and the overall drag coefficient was calculated using a blended scheme as the phase fraction weighted average of the different drag coefficients for each phase. The effect of volume fraction was also considered on the drag force unlike previous studies which used Schiller-Naumann drag model without considering the effect of volume fraction [41].

For simplicity, the dispersed phase was assumed to be mono-dispersed and the Sauter mean diameter was estimated using the unified correlation proposed by Sinha *et al* [9] (Eq 21)

$$\frac{d_{32}}{\sqrt{\frac{\sigma}{\Delta\rho g}}} = 1.197 * 10^3 \left(\frac{Af^4 \rho_c}{g\sigma}\right)^{-0.191} * \left(\frac{d_a \rho_c \sigma}{\mu_c^2}\right)^{-0.289} * \left(\frac{\rho_c \sigma^4}{\psi \mu_c}\right)^{0.110} * \left(\frac{\Delta\rho}{\rho_c}\right)^{-0.533} * \left(\frac{\mu_d}{\mu_c}\right)^{0.299} * \left(\frac{h_c}{d_a}\right)^{-0.591} * (1 + R)^{-0.009} \quad (21)$$

The surface tension force $\mathbf{F}_{S,k}$ was added to the model as given in Eq 22

$$\mathbf{F}_{S,k} = \kappa * \sigma * (\alpha_{disp.} * \nabla \alpha_{cont.} - \alpha_{cont.} * \nabla \alpha_{disp.}) \quad (22)$$

where κ is the surface curvature and is given in Eq 23

$$\kappa = -\nabla \cdot (\hat{\mathbf{n}} \cdot \vec{\mathbf{Sf}}) \quad (23)$$

In Eq 23, $\hat{\mathbf{n}}$ is the unit normal and $\vec{\mathbf{Sf}}$ is the area vector of a particular face.

In previous studies reporting holdup simulations via Eulerian multiphase models in PDDCs, the surface tension force was not included in the momentum equations. In this study, this force was added to make our simulations more robust and realistic.

The next force considered in the momentum equations is the virtual mass force. This force was expressed by Eq 24

$$\mathbf{F}_{vm,i} = C_{vm} * \alpha_{disp.} * \rho_{cont.} * \left(\frac{D\mathbf{U}_{cont}}{Dt} - \frac{D\mathbf{U}_{disp}}{Dt}\right) \quad (24)$$

In this study, the droplet Reynold's number remained below 1000, and the spheres were regarded as rigid with an immobile surface. Therefore, the value of C_{vm} was taken as 0.5 [25].

The lift force was assumed to have negligible effect on the dispersed phase holdup as argued by Bardin-Monnier *et al* [36] for Lagrangian simulations of PDDCs, and therefore it was omitted in the present study.

3.2. Turbulent modeling

Correct turbulence modeling is very important in an actual PDDC as it significantly influences the hydrodynamics of the system. Turbulence increases the interfacial area among the continuous and dispersed phases by generating eddies of a wide range of length scale starting from the smallest scale Kolmogorov eddies to larger scale energy containing eddies. These eddies interact with the droplet interfaces and enhance their breakage by transferring the turbulent kinetic energy from the continuous phase. These phenomena are typically explained by models like the k-epsilion, k-omega or k-omega-SST which will now be discussed.

Before, describing the turbulence models, let us define first the characteristic Reynold's number as per Eq 25 to confirm the presence of turbulence in our system

$$Re_c = \frac{d_h A * f}{d.c * v_c * \text{free area}} \quad (25)$$

where d_h is the hydraulic diameter defined as the ratio of the cross-sectional area to the wetted perimeter, $A*f$ is the product of pulsing amplitude (A) and pulsing frequency (f), known as the pulsation intensity or pulsing velocity, d.c. is the percentage duty cycle (d.c) and v_c is the continuous phase kinematic viscosity. As the pulsing velocity is much greater than the superficial velocity, the superficial velocity can be neglected in the Reynold's number estimation.

In our case, we found out that starting from lowest to highest value of superficial velocity and pulsation intensity, the characteristic Reynold's number evolved from 5112 to 10224 (>2100) indicating turbulence in our system. Two different turbulence models were implemented. In the VOF method, as a common shared velocity is used between the phases, the standard k-epsilon

turbulence model was used to account for the *effective phase* at higher superficial phase velocities (i.e. 0.314, 0.375 cm/s). However, since this model overpredicts the dispersed phase holdup at lower velocities (i.e. 0.125, 0.25 cm/s), the k-omega-SST turbulence model was used at lower velocities. In the hybrid multi-fluid-Eulerian method, only the standard k-epsilon turbulence was used to account for the entire velocity range only in the continuous phase in line with the study of Saini *et al* [39] for modeling the stage holdup in PDDCs using Eulerian multiphase models. To estimate the turbulent viscosity (μ_T) in the momentum equations for both models (Eq 4 & 15), the standard k-epsilon and k-Omega-SST models were implemented as described in sections 3.2.1 and 3.2.2, respectively.

3.2.1. Standard k-epsilon model

The standard k-epsilon model proposed by Launder and Spalding [66] was implemented using the set of equations described below.

3.2.1.1. k-equations

The continuous phase k-equation for the VOF and multi-fluid-Eulerian methods was expressed by Eq 25 and 26, respectively:

$$\frac{\partial \rho k}{\partial t} + \nabla \cdot (\rho \mathbf{U} k) = \nabla \cdot \left[\left(\mu + \frac{\mu_T}{\sigma_k} \right) \nabla k \right] + G + \rho \epsilon \quad (25)$$

$$\frac{\partial \rho_{\text{cont}} k}{\partial t} + \nabla \cdot (\rho_{\text{cont}} \mathbf{U}_{\text{cont}} k) = \nabla \cdot \left[\left(\mu_{\text{cont}} + \frac{\mu_T}{\sigma_k} \right) \nabla k \right] + G + \rho_{\text{cont}} \epsilon \quad (26)$$

where ρ and μ in Eq 25 refer to the effective phase and were calculated by Eq 8 and Eq 9, respectively. In both equations, k , G and ϵ are the turbulent kinetic energy, the turbulent kinetic energy generation term and the turbulent dissipation rate, respectively.

3.2.1.2. Epsilon equations

The equations for epsilon and the turbulent energy dissipation rate in the VOF and hybrid multi-fluid Eulerian method were given by Eq 27 and 28, respectively:

$$\frac{\partial \rho \epsilon}{\partial t} + \nabla \cdot (\rho \mathbf{U} \epsilon) = \nabla \cdot \left[\left(\mu + \frac{\mu_T}{\sigma_\epsilon} \right) \nabla \epsilon \right] + C_{1\epsilon} \frac{\epsilon}{k} G - C_{2\epsilon} \rho \frac{\epsilon^2}{k} \quad (27)$$

$$\frac{\partial \rho_{\text{cont}} \epsilon}{\partial t} + \nabla \cdot (\rho_{\text{cont}} \mathbf{U}_{\text{cont}} \epsilon) = \nabla \cdot \left[\left(\mu_{\text{cont}} + \frac{\mu_T}{\sigma_\epsilon} \right) \nabla \epsilon \right] + C_{1\epsilon} \frac{\epsilon}{k} G_{\text{cont}} - C_{2\epsilon} \rho_{\text{cont}} \frac{\epsilon^2}{k} \quad (28)$$

3.2.1.3. Turbulent parameters in k-epsilon model

In the k-epsilon model for both methods, G was modeled by Eq 29 and Eq 30 for the VOF and hybrid multi-fluid Eulerian methods, respectively:

$$G = \mu_T \left[\nabla \mathbf{U} : \{ \nabla \mathbf{U} + \nabla \mathbf{U}^T \} - \frac{2}{3} * (\nabla \mathbf{U})^2 \right] - \frac{2}{3} \rho k \nabla \mathbf{U} \quad (29)$$

$$G_{\text{cont}} = \mu_T \left[\nabla \mathbf{U}_{\text{cont}} : \{ \nabla \mathbf{U}_{\text{cont}} + \nabla \mathbf{U}_{\text{cont}}^T \} - \frac{2}{3} * (\nabla \mathbf{U}_{\text{cont}})^2 \right] - \frac{2}{3} \rho_{\text{cont}} k \nabla \mathbf{U}_{\text{cont}} \quad (30)$$

The turbulent viscosity for both methods was computed Eq 31 and 32 as follows

$$\mu_T = C_\mu \rho \frac{k^2}{\epsilon}; \mu_T = C_\mu \rho_{\text{cont}} \frac{k^2}{\epsilon} \quad (31, 32)$$

In Eq 25-28, σ_k and σ_ϵ are the turbulent Prandtl numbers with values of 1 and 1.3, respectively, whereas the values of $C_{1\epsilon}$ and $C_{2\epsilon}$ in Eq 27 and 28 were taken as 1.44 and 1.92, respectively, The empirical constant, C_μ , in Eq 31 and 32 was taken at 0.09 [67].

In the momentum equation for both methods (Eq 4 and 15), the modified pressure, p^* , was expressed by Eq 33 and 34

$$p^* = p + \frac{2}{3} * \rho k; p^* = p + \frac{2}{3} * \rho_{\text{cont}} k \quad (33, 34)$$

3.2.2. k-Omega-SST models

The k-omega-SST model [68] was used in the VOF method to describe the dispersed phase holdup at lower superficial phase velocities (i.e. 0.125 and 0.25 cm/s). The k-omega-SST model relies on a combination of the standard k-epsilon and k-omega turbulence models and uses a blending function to switch from one model to the other. The standard k-epsilon model is suitable for the free stream, while the k-omega model is adapted to describe boundary regions near the walls [67]. However, the k-omega turbulence model is heavily dependent on the free stream turbulence and small change in k can bring a large change in the turbulent viscosity.

The transport equation for k in the k-omega-SST model was expressed by Eq 35

$$\frac{\partial \rho k}{\partial t} + \nabla \cdot (\rho \mathbf{U} k) = \nabla \cdot \left[\left(\mu + \frac{\mu_T}{\sigma_k} \right) \nabla k \right] + G - \beta^* k \omega \quad (35)$$

The transport equation for the specific dissipation rate of omega ($\omega = \frac{\epsilon}{C_\mu k}$) was given by Eq 36

$$\frac{\partial \rho \omega}{\partial t} + \nabla \cdot (\rho \mathbf{U} \omega) = \nabla \cdot \left[\left(\mu + \frac{\mu_T}{\sigma_\omega} \right) \nabla \omega \right] + \alpha S^2 - \beta \omega^2 + 2(1 - F_1) \sigma_{\omega 2} \frac{1}{\omega} \nabla k \cdot \nabla \omega \quad (36)$$

where F_1 is the blending function for switching from k-omega model to k-epsilon model, where $F_1=1$ produces the transport equation for omega (Eq 35) and $F_1=0$ becomes the epsilon equation for the standard k-epsilon model (Eq 36). F_1 was expressed by Eq 37

$$F_1 = \tanh \left\{ \min \left[\max \left(\frac{\sqrt{k}}{\beta^* \omega y}, \frac{500\mu}{\rho y^2 \omega} \right), \frac{4\sigma_{\omega 2} k}{CD_{k\omega} y^2} \right] \right\}^4 \quad (37)$$

where y is the shortest distance to the next surface and was estimated by taking the ratio of the turbulent length scale ($L_T = \frac{\sqrt{k}}{C_\mu \omega} = \frac{k^{3/2}}{\epsilon}$) as 0.25 in the logarithmic region of the boundary layer, and $y=0$ towards the boundary layer edge.

$CD_{k\omega}$ is the cross-diffusion term that transforms the original transport equation for epsilon in the standard k-epsilon model into the k-omega model by incorporating F_1 . $CD_{k\omega}$ was expressed by Eq 38

$$CD_{k\omega} = \max \left(2\rho\sigma_{\omega 2} \frac{1}{\omega} \nabla k \cdot \nabla \omega, 10^{-10} \right) \quad (38)$$

In Eq 35 and 36, σ_k , σ_ω , α and β were estimated using Eq 39-42

$$\sigma_k = \sigma_{k1} F_1 + (1 - F_1) \sigma_{k2} \quad (39)$$

$$\sigma_\omega = \sigma_{\omega 1} F_1 + (1 - F_1) \sigma_{\omega 2} \quad (40)$$

$$\alpha = \alpha_1 F_1 + (1 - F_1) \alpha_2 \quad (41)$$

$$\beta = \beta_1 F_1 + (1 - F_1) \beta_2 \quad (42)$$

where $\sigma_{k1} = 0.85$; $\sigma_{k2} = 1$; $\sigma_{\omega 1} = 0.5$; $\sigma_{\omega 2} = 0.856$, $\alpha_1 = 0.55$, $\alpha_2 = 0.44$; $\beta^* = 0.09$.

To simulate the turbulent viscosity for free shear layers, the k-omega-SST model must be limited to the wall bounded flows. This was done by applying a second blending coefficient F_2 (Eq 43)

$$\mu_T = \frac{a_1 k}{\max(a_1 \omega, S F_2)} \quad (43)$$

where a_1 is the model constant with a value taken as 0.31, S is the shear strain, and F_2 was expressed as follows:

$$F_2 = \tanh \left[\left[\max \left(\frac{2\sqrt{k}}{\beta^* \omega y}, \frac{500\mu}{\rho y^2 \omega} \right) \right]^2 \right] \quad (44)$$

The turbulent energy generation term G in Eq 35 due to the mean velocity gradients was given by Eq 45

$$G = \min(\tau \nabla \mathbf{U}, 10\beta^* k \omega) \quad (45)$$

3.3. Computation details

The equations for the turbulent VOF and turbulent hybrid multi-fluid-Eulerian methods were solved using the open-source Computed Fluid Dynamics (CFD) OpenFOAM-v1906 code in 2-D. As reported in previous studies, the experimentally measured time averaged velocity for a PDDC in the azimuthal direction is nearly zero [37] and the flow inside the column is axisymmetric [29,31] allowing simulations in 2-D. Moreover, there are previous studies which reported useful results using a 2-D simulation in PDDC [41]. Therefore, in this study, the simulations were performed in 2-D. The discretization and meshing details of the governing equations, boundary conditions and computational domain are provided in the Supporting Information (sections S1-S4) and **Figure S2**. The phases were set at isothermal conditions; therefore heat transfer was not incorporated.

The VOF and hybrid multi-fluid-Eulerian method were implemented through the in-built interFOAM and multiphaseEulerFOAM solvers, respectively. The PIMPLE algorithm was used for both solvers as provided in **Figure S3**. Details on the calculations can be found in the Supporting Information (section S2).

4. Results and Discussion

4.1. Hydrodynamic regimes

We first investigated the hydrodynamic regimes in our experimental PDDC for the 3 M nitric acid- 30% tbp biphasic system evolving from the mixer-settler to the quasi-emulsion regime, and the results are presented in the **Figure 4**. Images for the mixer-settler, dispersion and quasi-emulsion regimes were measured at a superficial phase velocity of 0.125 cm/s and pulsation intensities at 0.936 cm/s, 1.56 cm/s and 1.872 cm/s, respectively. As seen from the images, layer formation is evident beneath the discs in the mixer-settler regime and a clear reduction the droplet size is observed when evolving from the mixer-settler to the dispersion and quasi-emulsion regimes. Apart from studying the visual change in the droplets, the dispersed phase holdup was also measured and it was found out that it reached a minimum from mixer-settler to pre-dispersion regime and then started to increase in the dispersion and quasi-emulsion regimes. Therefore, the pulsation intensity at transition regime was 1.25 cm/s for this study. This observation of transition regime through holdup measurement is in concurrence with the reported literature [7,8]. More detailed experimental observations with explanations can be found in a previous study [9].

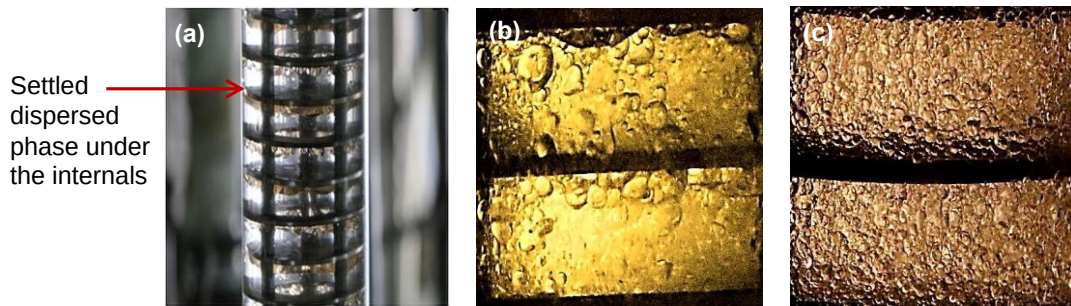


Figure 4: Regimes in the experimental PDDC with increasing pulsation intensity: (a) mixer-settler regime at a pulsation intensity of 0.936 cm/s, (b) dispersion regime at a pulsation intensity of 1.56 cm/s and (c) quasi-emulsion regime at a pulsation intensity of 1.872 cm/s. The superficial phase velocity was 0.125 cm/s.

With these results in hand, we computed the dispersed phase holdup in the operating regimes and at the onset of local and total flooding in our PDDC with 30% tbp-3 M nitric acid biphasic system by the VOF and hybrid multi-fluid-Eulerian methods using the interFOAM and multiphaseEulerFOAM solvers, respectively.

The simulations were performed from a minimum duration of 800 s to 1400 s to reach steady state. For both methods, the water-dodecane interface is observed to rise until steady state is reached. Next, we investigated the time-evolution of the dispersed phase holdup using both methods. Starting our analysis by the VOF method implemented with the interFOAM solver, irrespective of the superficial phase velocity, the system reaches steady state after 200-400 s for pulsation intensities in the range 0.936-1.25 cm/s that denotes a mixer-settler regime and the start of the dispersion regime (i.e. pre-dispersion) (**Figure 5**). At higher pulsation intensities (range 1.56-1.872 cm/s) encompassing a transition to dispersion and quasi-emulsion regimes, the system reaches steady state roughly after 600 s. The longer time required to reach steady state in the dispersion and quasi-emulsion regimes can be explained by a higher level of droplet breakage and coalescence resulting in slowest droplet stabilization compared to the mixer-settler and pre-dispersion regimes owing to the much higher turbulence in the column.

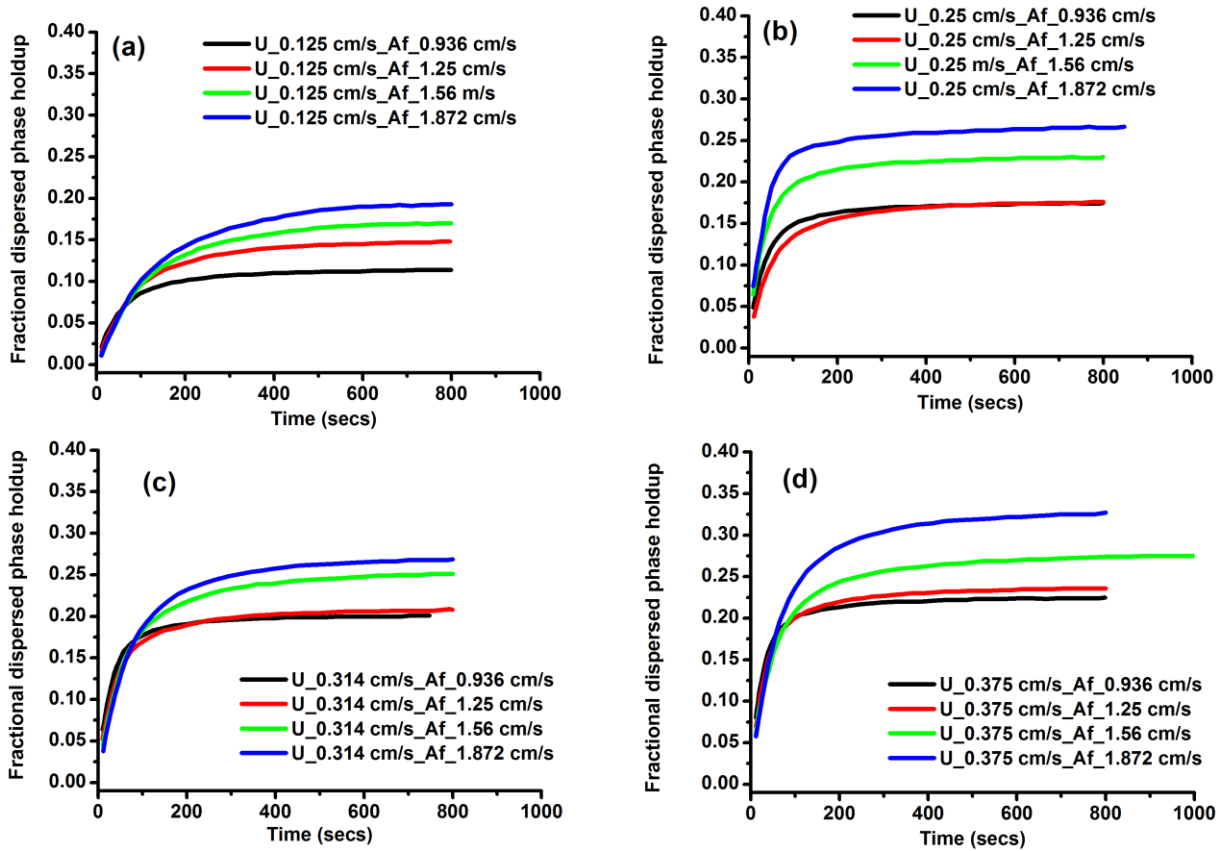


Figure 5. Time-evolution of the dispersed phase holdup for pulsation intensities in the range 0.936-1.872 cm/s at a superficial phase velocity of (a) 0.125 cm/s, (b) 0.25 cm/s, (c) 0.314 cm/s and (d) 0.375 cm/s for the VOF method implemented with the interFOAM solver.

To substantiate this, we simulated the initial settling time of the dispersed phase at zero to highest pulsation intensity of 1.872 cm/s at a phase superficial velocity of 0.375 cm/s. The simulated volume fractions were rescaled from 0-0.4 for better visualization of the dispersed phase in the PDDC. The results are given in **Figure 6**. By increasing the pulsation intensity, the settling time for the dispersed phase increases monotonically from 25-55 s. This shows that by increasing the pulsation, the droplets break into smaller ones resulting in a reduced settling velocity and increased settling time that ultimately delays the time to reach steady state.

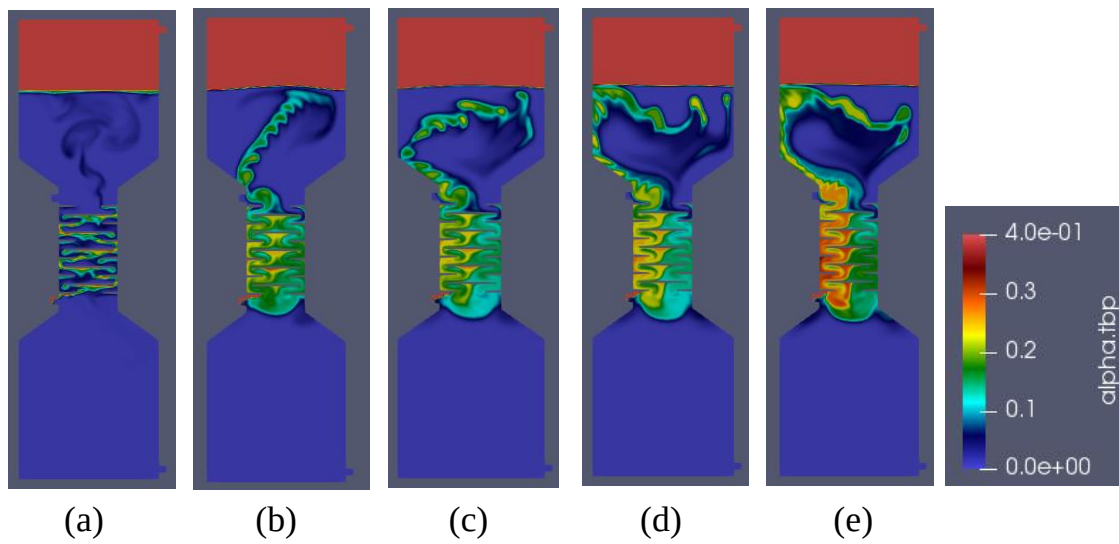


Figure 6. Simulated settling time of the dispersed phase at the interface at various pulsation intensities (Af) (a) 25 s; Af-0 cm/s (b) 40 s; Af-0.936 cm/s (c) 45 s Af-1.25 cm/s (d) 50 s; Af-1.56 cm/s (e) 55 s Af-1.872 cm/s; phase superficial velocity-0.375 cm/s.

Our analysis was extended next to the hybrid multi-fluid Eulerian model implemented with the multiphaseEulerFOAM solver (**Figure S4**). At lower superficial phase velocities (range of 0.125-0.25 cm/s), the time to reach steady state at lower pulsation intensities (0.936-1.25 cm/s) falls into the range 300-600 s (**Figure S4a-b**) that is slightly delayed compared to that obtained with the VOF method. We can attribute this difference to the inclusion of the drag force in the hybrid multi-fluid Eulerian model, whereas surface tension force is the main contributor to interfacial surface generation in the VOF method. However, at higher superficial phase velocities (0.314-0.375 cm/s) and lower pulsation intensities (0.936-1.25 cm/s), the time to reach steady

state is about 400 s (**Figure S4c-d**) which is comparable to that obtained in VOF simulations. The similar times obtained by both methods can be explained by an increase of inertial forces at higher velocities that promotes the shear force on the droplets and results in fast attainment of uniform droplet size distribution. Finally, at higher pulsation intensities in the range 1.56-1.872 cm/s (i.e. dispersion and quasi-emulsion regimes), regardless of the superficial phase velocity, the time to reach steady state is longer than 600 s that is also comparable to that simulated with the VOF method. Overall, this body of results point out that inertial forces play a major role in bringing the system towards steady state, and these forces are well captured using both methods.

4.2. Prediction of dispersed phase holdup

The dispersed phase holdup predicted by the VOF and hybrid multi-fluid Eulerian methods was compared to experimental holdup values as a function of the operation conditions (**Figure 7**). For turbulent VOF simulations using the k-epsilon model, the predicted dispersed phase holdups at higher velocities (range of 0.314-0.375 cm/s) match closely the experimental values with an average error (AARE) of only 5.5% (**Figure 7a**). The dispersed phase holdup increases monotonically with the pulsation intensity which can be explained by a decrease of the droplet size and settling velocity. However, at lower superficial phase velocities (0.125-0.25 cm/s), predictions with the k-epsilon turbulence model deviate from experimental values which can be attributed to enhanced wall effects. By correcting the turbulence occurring near the wall regions using the k-omega-SST model, the prediction level of the dispersed phase holdup improves drastically at 0.25 cm/s (**Figure 7c**). However, at 0.125 cm/s, the predicted dispersed phase holdup is unsatisfactory. Therefore, other turbulence models with lower Reynold's number may be attempted.

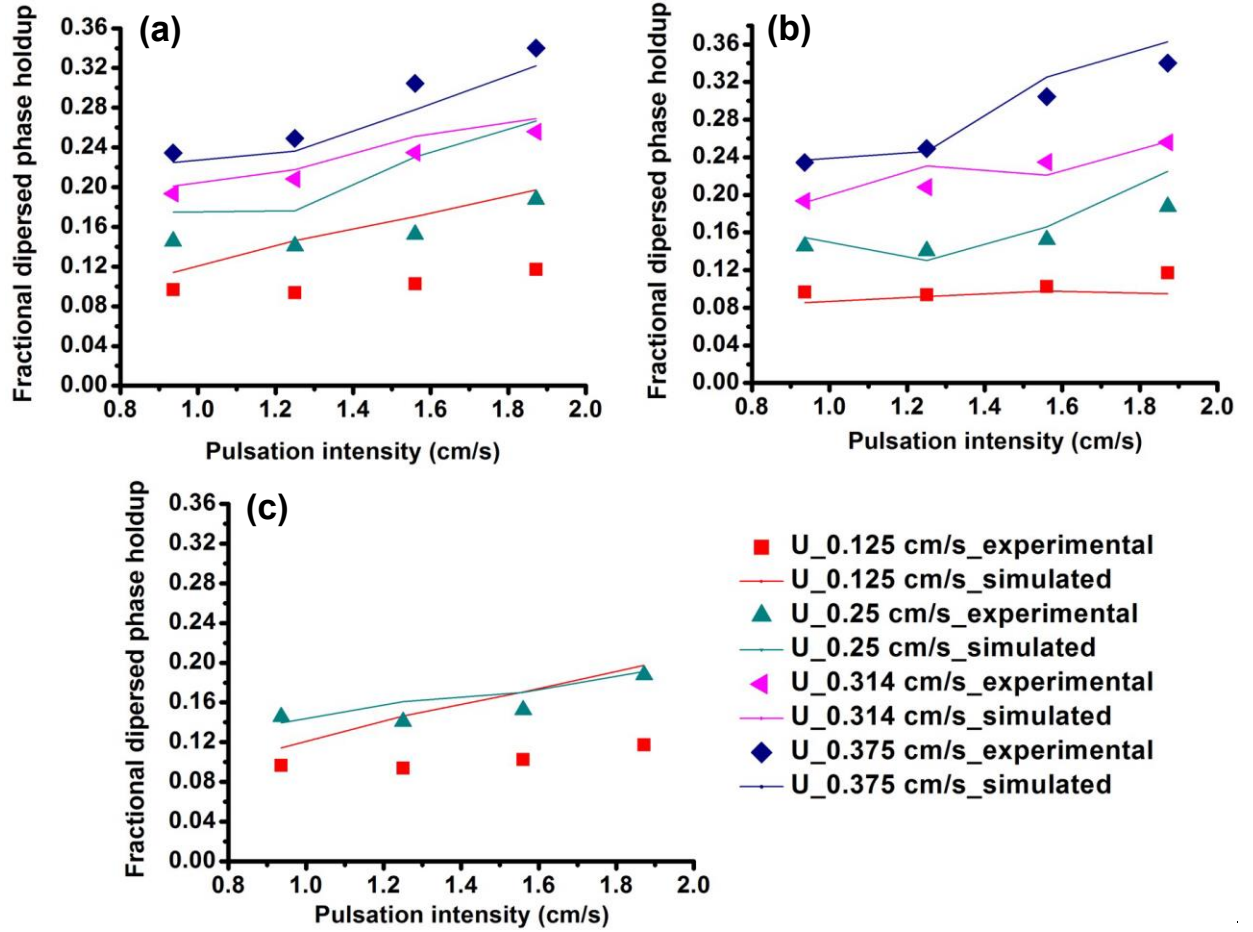


Figure 7. Experimental and predicted trends of the dispersed phase holdup as a function of the pulsation intensity predicted with (a) VOF method implemented with k-epsilon turbulence model and interFOAM solver, (b) hybrid multi-fluid Eulerian method implemented with k-epsilon turbulence model and multiphaseEulerFOAM solver, and (c) VOF method implemented with k-omega-SST turbulence model and interFOAM solver, for superficial phase velocities in the range 0.125-0.25 cm/s. Experimental and predicted results are represented by dots and lines, respectively.

Next, the dispersed phase holdup was simulated using the hybrid multi-fluid Eulerian method (**Figure 7b**). In this method, since the drag force is included in the momentum equation, the dispersed phase droplet diameter must be injected to the equations. In our approach, the droplet diameters were supplied using the unified correlation reported by Sinha *et al* [9] (Eq 21). Using this correlation, the error in the Sauter mean diameter calculation was 16.5%. To further improve the prediction level of the prediction of the dispersed phase holdup, multiple dispersed phase droplet diameters were attempted (at least 3-4 for each simulation) at four superficial phase

velocities (0.125, 0.25, 0.314 and 0.375 cm/s) and variable pulsation intensities. In all cases, after carrying out the trial-and-error method, the optimum dispersed phase droplet diameters which gave the best holdup predictions were selected. Moreover, the calculated dispersed phase droplet diameters fall within the average absolute relative error of the prediction ($\pm 16.54\%$) (**Figure S5**) which confirmed the applicability of the correlation used in this study. With this approach, an AARE% of 7.1% was obtained for the dispersed phase holdup prediction. It is to be noted that to treat larger droplets separately, the population balance model might be a good choice which was not in the scope of this study and that may lead to increased computational cost. Nevertheless, we also tried to predict the wobbling of a larger droplet (4-mm diameter), characterized by oscillations in the path of while rising, by the hybrid Eulerian method with a constant diameter assumption to check its efficiency. The plot of the droplet positions with respect to the axial and radial direction is given in **Figure S6**. As can be observed, the droplet wobble is well predicted along the axial direction. Therefore, the constant diameter model can be used in our case.

We explored the effect of the pulsation intensity on the dispersed phase droplet diameter (**Figure 8**). Regardless of the superficial velocity, the dispersed phase droplet diameter decreases sharply until the pulsation intensity of 1.25 cm/s due to a dominant effect of inertial forces, but the change is smaller between 1.25-1.872 cm/s that we qualitatively attribute to larger viscous forces and higher internal pressure. This trend matches earlier reported experimental observations [8,9,16].

Since the VOF and hybrid multi-fluid Eulerian methods can predict with high level of accuracy the dispersed phase holdup at the higher superficial phase velocities (i.e. 0.314 and 0.375 cm/s), we implemented both methods to predict the axial holdup at every stage in the column. The axial holdup is a relevant parameter as is directly related to the axial mass transfer and thus the HTU of the column. The mean axial holdup was calculated between every doughnut and disc along the column height for pulsation intensities in the range 0.936-1.872 cm/s. Both methods provide a similar trend for the axial holdup (**Figure 9, Figure S7**). The mean axial

holdup increases first with the column height until the 9th stage and decreases further in the final stage for a superficial phase velocity of 0.314 cm/s and pulsation intensities in the range 0.936-1.25 cm/s denoting mixer-settler and the start of the dispersion regime (**Figure 9a**). This observation suggests the initial formation of larger droplets at the bottom of the column resulting in a lower holdup due to a higher settling velocity. As the droplets travel upwards, they break due to collision with the column internals and walls resulting in smaller droplets that reduce their settling velocity and in turn increase the axial holdup. At the last stage, just before entering the upper end-settler of the column, the area increases and phase separation starts that accelerates droplet coalescence reducing sharply the axial holdup. Interestingly, at pulsation intensities in the range 1.56-1.872 cm/s (dispersion and quasi-emulsion regime), the axial holdup increases until the 8th stage but decreases upwards. The presence of a maximum for the axial holdup can be explained by the generation of smaller droplets upon breakage that promotes the holdup. This also means that the coalescence probability increases at higher axial positions in the column resulting in the formation of larger droplets.

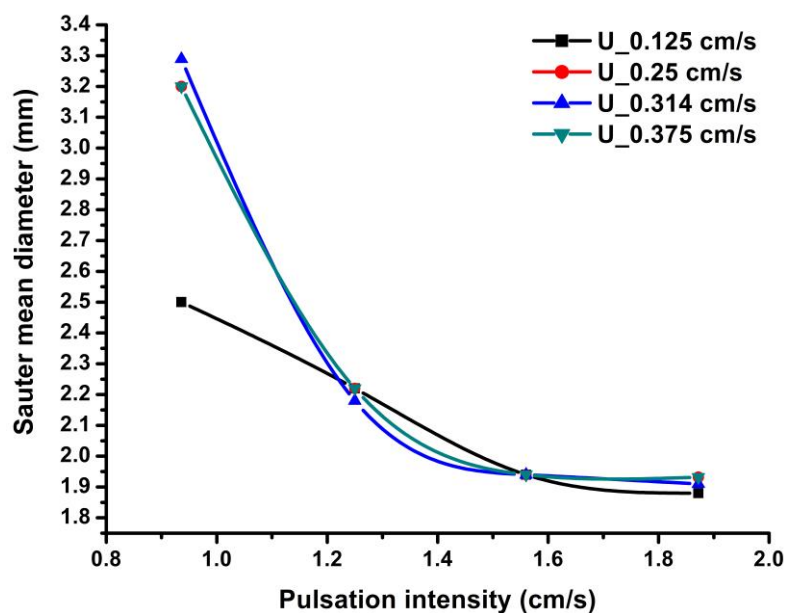


Figure 8. Effect of the pulsation intensity on the dispersed phase droplet diameter simulated by the hybrid multi-fluid Eulerian method. The curves are a guide to the eye.

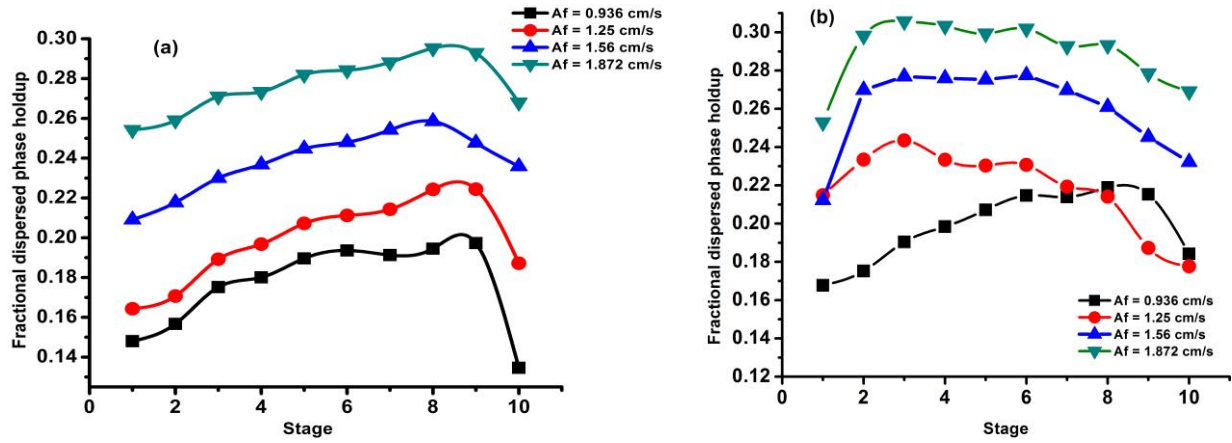


Figure 9. Evolution of the axial holdup with the column height at variable pulsation intensities predicted by the VOF method for a superficial phase velocity of (a) 0.314 cm/s and (b) 0.375 cm/s. The curves are a guide to the eye. Simulations using the hybrid multi-fluid Eulerian method are provide in the Supporting Information (**Figure S7**).

We next conducted simulations using the VOF method at a superficial phase velocity of 0.375 cm/s and a pulsation intensity of 0.936 cm/s (mixer settler regime) (**Figure 9b**). The axial holdup displays a comparable trend as a function of the column height to that observed at 0.314 cm/s. However, at higher pulsation intensities in the range 1.56-1.872 cm/s, the axial holdup increases only until the 3rd stage, then becomes steady after until the 6th stage, and gradually decreases afterwards with the column height. We attribute the steady behavior to a more intense droplet breakage at higher pulsation intensity that counters coalescence at higher superficial phase velocity and therefore the droplet diameter keeps almost unchanged resulting in a steady axial holdup. On the other hand, we attribute the declining trend after the 6th stage to an enhanced droplet coalescence resulting in a larger size and higher settling velocity.

With these results, we investigated the radial velocity profiles for the first 4 stages of the column height to gain insight into potential recirculation zones inside the column (**Figure 10**). The flow enters the discs from the annular area bedside in stages 2&4 and exits from the doughnut aperture in stages 1&3 (**Figure S8**). The velocity decreases sharply from the column wall to the center in stages 1&3 (**Figure 10a,c**). In contrast, in stages 2&4, the velocity increases from the column wall to the doughnut aperture and decreases towards the center of the column (**Figure**

10b,d). These observations can be explained by an increase of the flow area while exiting through the annulus besides the discs. This reduces the velocity and, while entering the doughnut area, the flow area decreases that promotes in turn the velocity. As a result, recirculation zones appear beneath the discs that are indicated by a red arrow in **Figure 11**. The zones simulated by the VOF method are slightly larger than those simulated by the hybrid multi-fluid Eulerian method (**Figure S9**). The larger recirculation zones in the former case might be ascribed to a slightly higher turbulent kinetic energy together with a faster dissipation rate due to viscous forces (**Figure S10**).

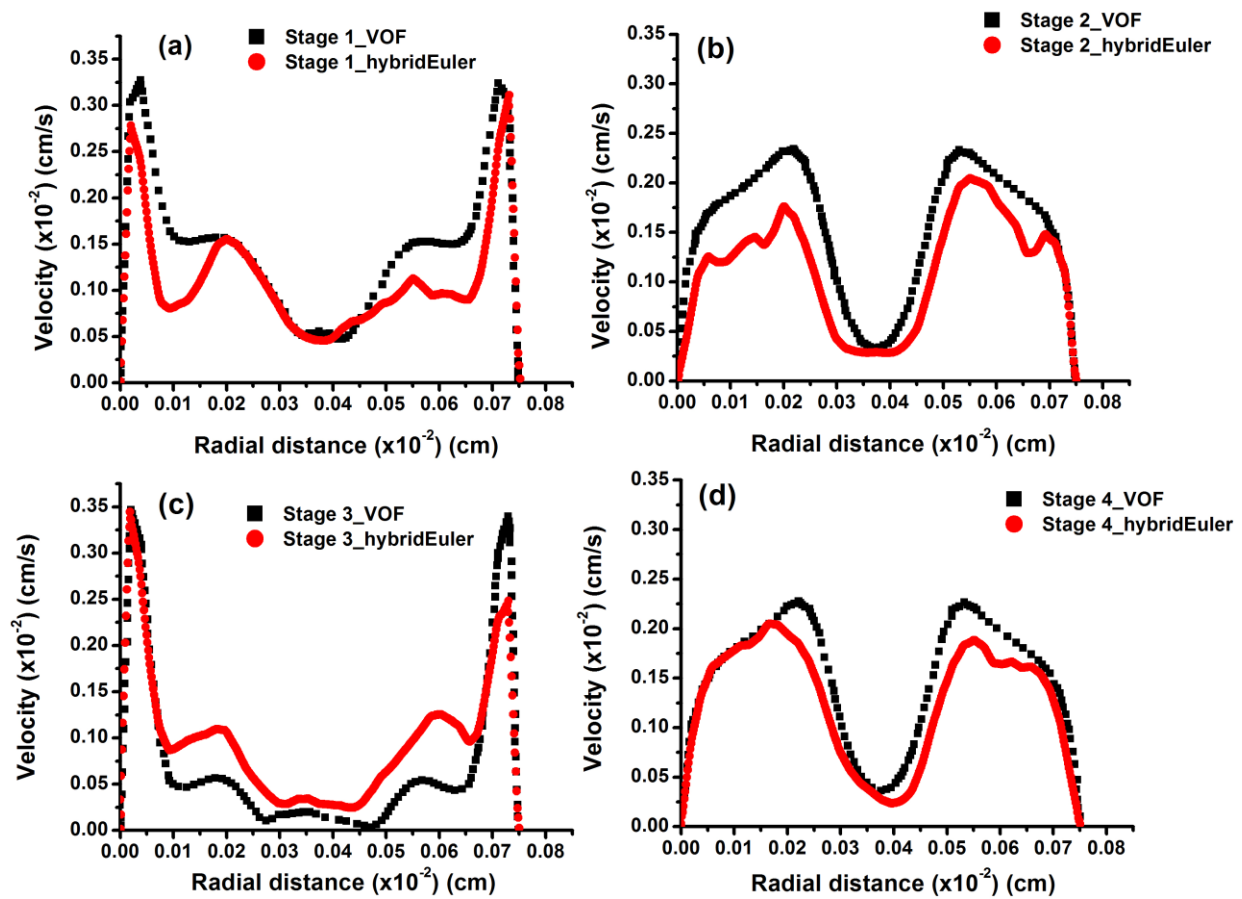


Figure 10. Radial velocity profiles computed by the VOF and hybrid multi-fluid Eulerian methods for the first 4 stages (S1-0.395 m; S2-0.406 m; S3-0.42 m; S4-0.431; S4-0.445 m) of the column at a superficial phase velocity of 0.314 cm/s and a pulsation intensity of 1.56 cm/s.

We also simulated the volume fraction fields of the dispersed phase by both methods at a superficial phase velocity of 0.375 cm/s and a pulsation intensity of 1.872 cm/s (**Figure 12**). Both methods predict the holdup with a comparable accuracy, but the length scales are better resolved

with the hybrid multi-fluid Eulerian method. It is worth mentioning that traditional Eulerian multiphase simulations do not resolve properly length scales due to the assumption of interpenetrating continuum. However, by incorporating an extra volume fraction equation for each phase along with the surface tension force, the hybrid multi-fluid Eulerian method can simulate the length scale with much higher resolution. To achieve the same length scale resolution with the VOF method, the mesh needs to be finer which increases the computational cost. Since the length scale resolution is not the aim of this work, the VOF method was used in this study to simulate the dispersed phase holdup. In addition, using the VOF method, the grid size is comparable to the smaller drops (diameter~1.5 mm) but smaller than the larger drops (diameter~3.2 mm). Therefore, the smallest drops are not fully resolved. However, despite the smearing of individual droplet interfaces, we achieved good predictions for the average dispersed phase holdup over the extraction section which is at a much larger length scale than the droplet scale. To resolve the smallest droplet with the VOF method, the mesh requires to be finer which increases the computational cost. Since the aim of this study was to compare the VOF method with the hybrid Eulerian method, the grid size was kept at the same size to simulate the dispersed phase holdup.

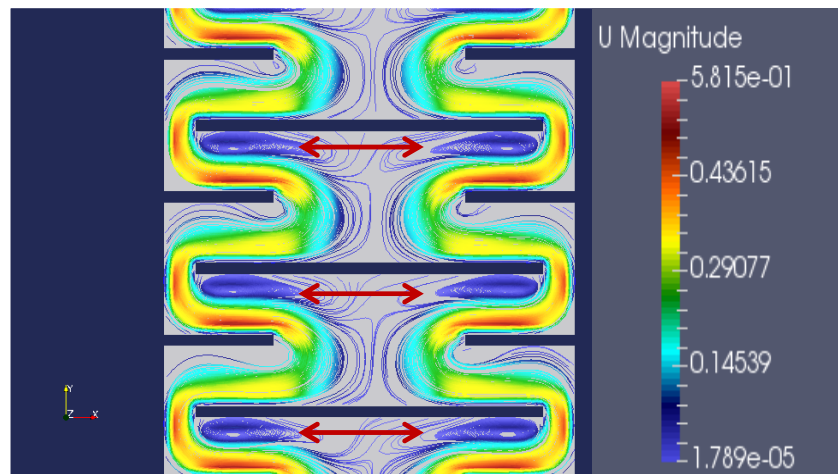


Figure 11. Recirculating zones (indicated by red arrows) simulated by the VOF method at a superficial phase velocity of 0.314 cm/s and pulsation intensity of 1.56 cm/s. The profiles simulated with the hybrid multi-fluid Eulerian method are provided in **Figure S9**.

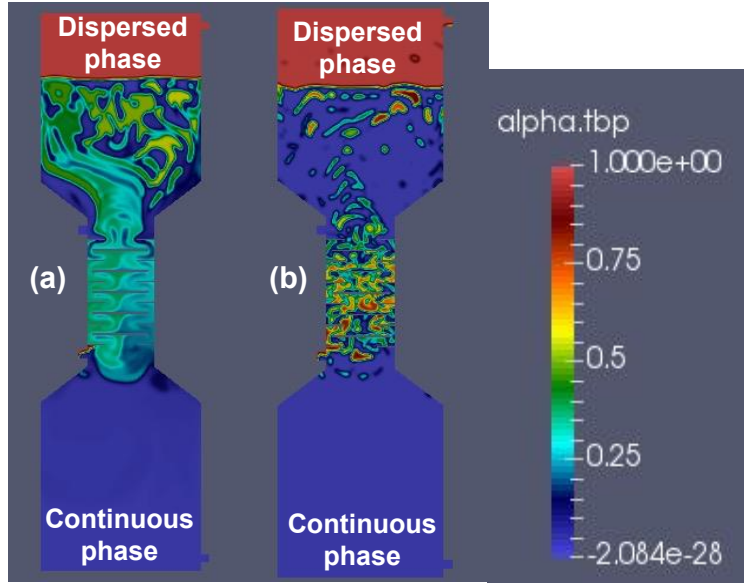


Figure 12. Volume fraction fields simulated by the (a) VOF and (b) hybrid multi-fluid Eulerian methods at a superficial phase velocity of 0.375 cm/s and pulsation intensity of 1.872 cm/s.

4.3. Prediction of flooding conditions

Flooding is a central phenomenon in PDDCs as it determines the maximum operable range for liquid-liquid extraction. As a rule, a PDDC can approach flooding in two ways. First, flooding can occur by an excessive droplet coalescence at a given pulsation intensity driven by a gradual increase of the superficial phase velocity. Second, flooding can occur by an excessive droplet dispersion due to a very high pulsation intensity at a given superficial phase velocity that breaks the droplets in smaller sizes that block the flow of the phases. This results in an accumulation of the dispersed phase in the column which ultimately exits from the continuous phase outlet. Although there are experimental studies related to the flooding conditions in PDDCs [17,18], to our knowledge no dedicated studies have been reported so far to predict and understand flooding in PDDCs. To fill this gap, flooding conditions were simulated with the VOF and hybrid multi-fluid Eulerian methods at the pulsation intensity of 1.56 cm/s (dispersion regime) in the phase superficial velocity range from 0.352 to 0.595 cm/s.

The experimental results of the onset of flooding and total flooding are validated in terms of flooding holdup (**Figure 13**). In the local flooding condition, the dispersed phase starts to form

globules due to local coalescence of droplets that results in a sharp increase in the dispersed phase holdup. This condition prevails until inter-droplet coalescence is enhanced excessively, and the dispersed phase holdup is again observed to shoot-up to a high value, i.e. $\sim 41\%$ of the volume of the active section of column. At this condition, the column is no longer operable and hence this condition was used to define the onset of total flooding.

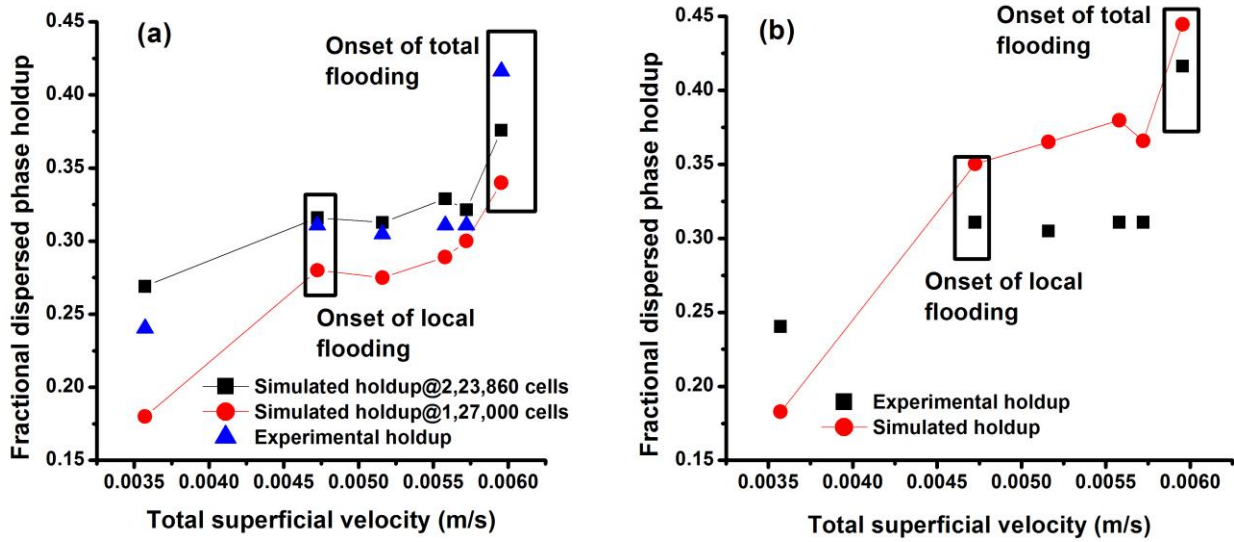


Figure 13. Validation of flooding conditions with (a) VOF and (b) hybrid multi-fluid Eulerian methods at a pulsation intensity of 1.56 cm/s.

Figure 14 plots the predicted volume fraction fields at the onset of local and total flooding using both methods. Using the VOF method, the length scales are not resolved properly using a mesh of 1,27,000 cells (**Figure 14a**), and the simulations of dispersed phase holdup at flooding condition do not reproduce satisfactorily the experimental values (**Figure 13a**). By increasing the number of cells to 2,23,860 one can notice the immediate enhancement of the turbulent length scale resolution (**Figure 14b**) that improves drastically the prediction of the onset of local and total flooding (**Figure 13a**). However, the error was still around 10%. The hybrid multi-fluid Eulerian method predicts the onset of local flooding with slight less accuracy than the VOF method but predicts accurately the onset of total flooding with an error $\sim 5\%$.

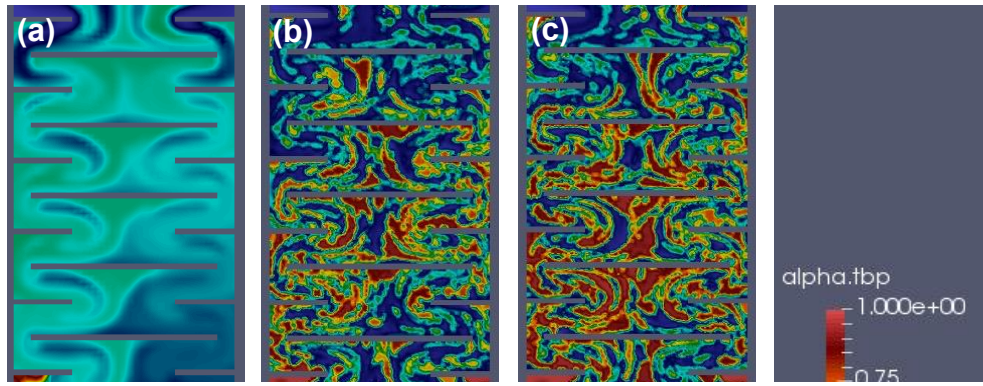


Figure 14. Volume fraction fields simulated with the VOF method for (a) onset of local flooding with 1,27,000 cells, (b) onset of local flooding with 2,23,860 cells, and (c) onset of total flooding with 2,23,860 cells. The volume fraction fields simulated with the hybrid multi-fluid Eulerian method are provided in **Figure S11**.

The velocity streamlines were also analyzed to visualize the recirculating zones arising at different velocities [**Figure 15a1,b1**]. At the onset of local flooding [**Figure 15a1**], along with the recirculation zones under the discs, relatively weaker recirculation zones are present (indicated with a black arrow) under the doughnut aperture, and grow in every stage for the onset of total flooding [**Figure 15a2**] (denoted by black arrow). We attribute the formation of these auxiliary recirculation to the observed sharp increase of the dispersed phase holdup at the onset of flooding and, as they grow stronger, the dispersed phase holdup increases heavily leading to a condition total flooding.

A similar analysis was carried out using the hybrid multi-fluid Eulerian method for the volume fraction fields at the onset of local and total flooding (**Figure S11**). Lumps of the dispersed phase are clearly visible (**Figure S11a**) that are responsible for the onset of flooding resulting in the initial increase in the holdup (**Figure 13b**). Excess accumulation of the dispersed phase is also visible (**Figure S11b**) that is responsible for the onset of total flooding in the column. It is worth mentioning that with traditional Eulerian multiphase modeling, the above resolution in terms of turbulent length scale resolution can hardly be achieved. The predictions by the hybrid multi-fluid Eulerian method are superior because of the incorporation of the surface

tension force in the momentum equation and the scalar transport equation for the volume fraction in each phase.

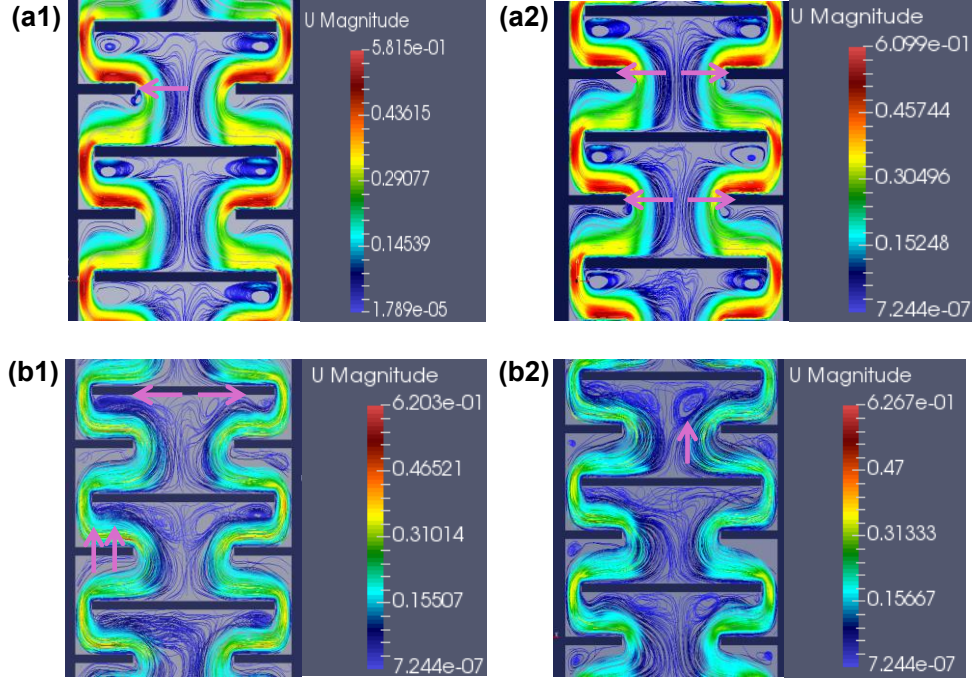


Figure 15. Recirculation zones simulated with the VOF method (top) and hybrid multi-fluid Eulerian approach (bottom) at the onset of (a1, a2) local flooding and (b1, b2) total flooding.

Subsequently, we analyzed the recirculation zones for the onset of local and total flooding (**Figure 15**). Using the VOF method, at the onset of local flooding, small auxiliary recirculation zones are observed under the doughnut aperture (denoted by black arrows) along with the recirculation zones under the discs [**Figure 15a1**]. In contrast, using the hybrid multi-fluid Eulerian method, multiple recirculation zones appear at the central region just under the discs of the column (denoted by red arrows) [**Figure 15a2**]. The dispersed phase holdup is quite high in the central region under the discs. At the onset of total flooding, with the VOF method, the auxiliary recirculation zones (denoted by black arrows) are more frequent under the doughnut aperture [**Figure 15b1**], whereas with the hybrid multi-fluid Eulerian method, the recirculation zones in the central region under the discs (denoted by red arrows) are stronger compared to the onset of flooding [**Figure 15b2**]. As a result, large accumulation of dispersed phase is observed

in the recirculation zones. Overall, this body of results point out that, due to recirculation, the dispersed phase holdup increases to such a high value that the column enters the state of total flooding.

Conclusions

A 2-D multiphase simulation study was conducted in a 7.5-cm diameter PDDC at four levels of superficial phase velocity and pulsation intensity. The simulations were conducted for the first time with the VOF and the hybrid multi-fluid Eulerian methods and the dispersed phase holdups were validated using experimental data on a laboratory scale PDCC. At higher superficial phase velocity (range 0.314-0.375 cm/s), both methods implemented with the k-epsilon turbulence model can predict the dispersed phase holdup with high accuracy. However, the VOF method performs better in terms AARE%. At lower superficial phase velocity (range 0.125-0.25 cm/s), the VOF method implemented with k-epsilon turbulence model does not provide a satisfactory prediction of the dispersed phase holdup. However, at a superficial phase velocity of 0.25 cm/s, the VOF method provides good prediction when implemented with the k-omega-SST turbulence model by including wall effects. Nevertheless, at the lowest superficial velocity (0.125 cm/s), the VOF method still provides poor accuracy. Therefore, it is necessary to develop alternative turbulence models for the VOF method with an improved treatment of wall effects.

The hybrid multi-fluid Eulerian method reproduces well the experimental trends for the dispersed phase holdup regardless of the superficial phase velocity. However, the input dispersed phase droplet diameter needs to be optimized at each level of superficial velocity and pulsation intensity which makes the calculations expensive. Therefore, the VOF method is recommended at higher superficial phase velocities to predict the dispersed phase holdup.

The axial dispersed phase holdup shows similar trends for both methods. These results clearly point out that by increasing the pulsation intensity, the coalescence probability increases due to a higher holdup. Higher superficial phase velocity increases the inter-droplet coalescence even more, and the axial holdup exhibits its maximum at the bottom regions of the column.

However, at higher pulsation intensities, the dispersed phase holdup shows almost a constant value region in the middle section of the column due to larger droplet breakage. The axial velocity decreases while exiting from the annulus beside the discs and increases while exiting from the doughnut aperture resulting in recirculation zones. These zones are more prominent using the VOF method because of a higher turbulent kinetic energy and dissipation rate.

To predict the onset of flooding, the mesh needs to be finer using the VOF method, but the onset of local flooding is better predicted. The error for the onset of total flooding is ~10%. Using the hybrid multi-fluid Eulerian method, the onset of total flooding is predicted with higher accuracy owing to the inclusion of additional recirculation zones in the central region just beneath the discs. In terms of industrial operation, the prediction of the onset of local flooding should be enough to set the operation limits. Therefore, the VOF method with a finer mesh is still recommended since, for the hybrid multi-fluid Eulerian method, the dispersed phase droplet diameter needs to be optimized at each operating condition as the dispersed phase holdup is very sensitive to the dispersed phase droplet size.

Associated Content

Supporting Information.

The Supporting Information contains the caption of the figures and tables. In addition, it is divided into four major sections. S1 contains the description of the discretization schemes in which S1.1, 1.2, 1.3 and 1.4 describes the temporal, gradient, divergence and Laplacian schemes. S2 contains the description of the boundary conditions in which S2.1, 2.2, 2.3 and 2.4 describes the inlet/outlet, top, bottom and wall boundary conditions, respectively. S3 describes the computational domain and S4 describes the calculation algorithms.

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