



**HAL**  
open science

# Use of CFD for Pressure Drop, Liquid Saturation and Wetting Predictions in Trickle Bed Reactors for Different Catalyst Particle Shapes

Hanane Bouras, Yacine Haroun, Francisco Fortunato Bodziony, Régis Philippe, Pascal Fongarland, Frédéric Augier

► **To cite this version:**

Hanane Bouras, Yacine Haroun, Francisco Fortunato Bodziony, Régis Philippe, Pascal Fongarland, et al.. Use of CFD for Pressure Drop, Liquid Saturation and Wetting Predictions in Trickle Bed Reactors for Different Catalyst Particle Shapes. *Chemical Engineering Science*, 2022, 249, pp.117315. 10.1016/j.ces.2021.117315 . hal-03519911

**HAL Id: hal-03519911**

**<https://ifp.hal.science/hal-03519911>**

Submitted on 10 Jan 2022

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

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

1            ***Use of CFD for Pressure drop, liquid saturation and wetting***  
2            ***predictions in trickle bed reactors for different catalyst***  
3            ***particle shapes***

4            **Hanane Bouras<sup>1,2</sup>, Yacine Haroun<sup>1,\*</sup>, Francisco Fortunato Bodziony<sup>1</sup>, Régis Philippe<sup>2</sup>,**  
5            **Pascal Fongarland<sup>2</sup>, Frédéric Augier<sup>1</sup>**

6            **\* Corresponding author: [yacine.haroun@ifpen.fr](mailto:yacine.haroun@ifpen.fr)**

7            1 : IFP Energies Nouvelles, Rond-point de l'échangeur de Solaize, BP 3, 69360 Solaize, France

8            2 : Univ. Lyon, CPE Lyon, Univ. Claude Bernard Lyon 1, CNRS, Laboratoire de Catalyse, Polymérisation,  
9            Procédés et Matériaux (CP2M) UMR 5128, 43 Bd du 11 novembre 1918, 69616 Villeurbanne, France.

10            ***Abstract***

11            The characterization of hydrodynamics in Trickle-Bed-Reactors is a complex task due to  
12            the opacity of the medium. In particular, the determination of pressure drop, liquid hold-up,  
13            wetting of the catalyst surface and catalyst shape effect on these parameters is very important  
14            for optimal catalyst use and reactor operation. Measurements under industrial conditions are  
15            limited to indirect estimations, and direct measurement methods are limited to near-ambient  
16            conditions. In this context, the objective of the present article is to use Computational-Fluid-  
17            Dynamics to investigate pressure drop, liquid saturation and wetting efficiency in Trickle-Bed-  
18            Reactors and to improve existing correlations, with a special focus on the catalyst shape effect  
19            and wetting prediction.

20            The Volume-Of-Fluid approach was used to simulate two-phase flow through particle  
21            loadings of spherical, trilobe and quadrilobe-shaped particles. The numerical model was

22 validated against literature correlations in terms of pressure drop, liquid holdup and wetting  
 23 efficiency. The CFD model was then employed to explore two effects that does not reach out  
 24 a consensus in existing literature, i.e effects of particle shape and gas-phase velocity on  
 25 wetting efficiency. As a result, it was shown that CFD provides good predictions of pressure  
 26 drop and liquid saturation for different catalyst particle shapes, the achieved deviations  
 27 between CFD results and correlation estimations are below 20%. A new wetting efficiency  
 28 correlation is also proposed. This new correlation is able to predict wetting efficiency with a  
 29 precision of 6.99% for a wide range of liquid velocities (from 0.2 to 0.8cm/s) and gas velocities  
 30 (from 5 to 20cm/s) and three particle shapes.

31 **Keywords:** Trickle bed reactors; pressure drop; liquid hold up, wetting efficiency; catalyst shape; CFD;  
 32 volume of fluid method; multiphase reactor; hydrocarbon

### 33 ***Nomenclature***

$a_s$	packing specific area, $m^2/m^3$ , $\frac{a_p}{V_p}$
$a_p$	Particle surface area, $m^2$
$d_p$	particle diameter, $m$
$d_e$	Particle equivalent diameter, $m$ , $\frac{6V_p}{a_p}$
$d_h$	Hydrodynamic diameter, defined by Krischer and Kast, $m$ , $\left(\frac{16\varepsilon_B^3}{9\pi(1-\varepsilon_B)^2}\right)^{0.33} d_p$
$f$	Wetting Efficiency, -, $S_w/S_p$
$g$	Gravitational acceleration, $m/s^2$
$u_L$	Liquid phase velocity, $m/s$
$u_G$	Gas phase velocity, $m/s$
$V_{S,L}$	Liquid phase superficial velocity, $m/s$
$V_{S,G}$	Gas phase superficial velocity, $m/s$
$V_L$	Liquid volume, $m^3$
$V_R$	Reactor volume, $m^3$
$S_p$	Solid particles' surface, $m^2$
$S_w$	Wetted solid surface, $m^2$

### 34 **Greek Letters**

$\alpha_G$	Gas phase volume fraction
$\alpha_L$	Liquid phase volume fraction
$\delta$	Liquid film thickness, <i>m</i>
$\beta_L$	Liquid saturation, -, $\frac{V_L}{\varepsilon_B V_R}$
$\varepsilon_B$	Bed void fraction/porosity
$\mu_m$	Mixture dynamic viscosity, <i>Pa.s</i>
$\mu_G$	Gas phase dynamic viscosity, <i>Pa.s</i>
$\mu_L$	Liquid phase dynamic viscosity, <i>Pa.s</i>
$\rho$	Mixture density, <i>kg/m<sup>3</sup></i>
$\rho_G$	Gas phase density, <i>kg/m<sup>3</sup></i>
$\rho_L$	Liquid phase density, <i>kg/m<sup>3</sup></i>
$\sigma_L$	Surface Tension, <i>N/m</i>
$\theta$	Contact angle, °

### 35 Dimensionless numbers

$Fr_G$	gas phase Froude Number $\frac{V_{S,G}^2}{gd_p}$
$Fr_L$	liquid phase Froude Number $\frac{V_{S,L}^2}{gd_p}$
$Ga_G$	gas phase Galileo Number $\frac{d_p^3 \rho_G^2}{\mu_G^2}$
$Mo_L$	Liquid phase Morton number, $\frac{g \mu_L^4}{\sigma_L^3 \rho_L}$
$Re_G$	gas phase Reynolds Number $\frac{u_G \rho_G d_p}{\mu_G}$
$Re_L$	liquid phase Reynolds Number $\frac{u_L \rho_L d_p}{\mu_L}$
$Re'$	Modified liquid phase Reynolds Number $\frac{u_L \rho_L d_p}{\mu_L (1 - \varepsilon_B)}$

### 36 Abbreviations

TBR	Trickle-Bed Reactor
CFD	Computational Fluid Dynamics
VOF	Volume-Of-Fluid
CSF	Continuum Surface Force
CFL	Courant–Friedrichs–Lewy
PISO	Pressure-Implicit with Splitting of Operators
PRESTO	PREssure-STaggering-Option
HDT	Hydrotreatment
SSE	Sum of Squared Errors
GRG	Generalised Reduced Gradient

37

## 38 **1. Introduction**

39 Trickle-bed reactors (TBRs) are gas-liquid-solid continuous reactors, where gas and liquid  
40 flow co-currently downward through a fixed bed of solid catalyst particles. Parallel to fluids'  
41 flow, the reagents travel through the phases to react at the surface or inside the porous  
42 catalyst particles. Trickle-bed reactors owe their popularity to the unique advantages they  
43 offer for large volume processing in the chemical and petroleum industries. Indeed, TBRs are  
44 characterised with a good trade-off between (i) scalability, (ii) gas-liquid-solid interfacial  
45 contact (iii) mass, heat and momentum transports. Due to their relatively easy conception and  
46 operability, TBRs are the best technology for many industrial processes to perform  
47 hydrogenations (e.g. hydrocracking, hydrodesulfurization, hydrometallization, etc.) and are  
48 used as well in biochemical processes (e.g. Fischer-Tropsch (Nishizawa et al. 2014)) and  
49 wastewater treatment applications.

50 Several experimental works were conducted in the past four decades to characterise the  
51 hydrodynamic behaviour of trickle-bed reactors, leading to well-established hydrodynamic  
52 correlations for a wide range of operating conditions. Regarding two-phase pressure drop,  
53 available correlations can be classified into three main categories. The first category consists  
54 of correlations based on Lockhart and Martinelli (1949) who correlated  $\Delta P_{GL}$  to the following  
55 dimensionless numbers: (i)  $\chi^2 = \Delta P_L / \Delta P_G$ , (ii)  $\Phi_G^2 = \Delta P_{GL} / \Delta P_G$  and (iii)  $\Phi_L^2 = \Delta P_{GL} / \Delta P_L$ . This  
56 model was the foundation to many other experimental works (Larkins et al. (1961), Sato et al.  
57 (1973), Midoux et al. (1976), Rao et al. (1983), Tosun (1984), Ratman et al. (1993)).  
58 Nevertheless, the aforementioned dimensionless numbers ( $\chi, \Phi_G, \Phi_L$ ) require the estimation  
59 of single-phase pressure drops ( $\Delta P_G$  and  $\Delta P_L$ ) in two-phase flow conditions, which proved to  
60 be challenging. Therefore, to overcome this challenge, a second correlation group based on  
61 bed characteristics and operating conditions emerged. Indeed, Turpin and Huntington (1967)

62 suggested an independent variable  $Z$  ( $Z = Re_G^{1.167}/Re_L^{0.767}$ ) discussed and used by other  
63 authors (Specchia and Baldi (1977), Rao et al. (1983), Sai and Varma (1987)). In their  
64 expressions, these correlations share some common points such as using the void fraction to  
65 the power of 3 to account for bed porosity effect on pressure drop. Finally, due to the empiric  
66 nature of the aforementioned correlation categories, a phenomenological model was  
67 presented by Holub et al. (1992) for the low-interaction regime, assuming that the flow inside  
68 the reactor is analogous to a flow in a 1D slit. Al-Dahhan and Dudukovic (1995) later expanded  
69 on this work, by performing experiments at high pressures and widening the application range  
70 of the correlation, capable of predicting both pressure drop and liquid holdup. In an effort to  
71 investigate operating pressure effect on two-phase pressure drop, Larachi et al. (1991)  
72 proposed a correlation to estimate  $\Delta P_{GL}$  in the trickle flow regime. The authors were among  
73 the first to study the effect of high operating pressure on pressure drop. More recently, Boyer  
74 et al.(2007) proposed a new mechanistic model to predict the pressure drop and liquid  
75 saturation inside the TBR. This correlation accounts for the tortuosity of the liquid film and  
76 provides good predictions of the hydrodynamic behaviour inside TBRs. An effort was also  
77 made by Bansal et al. (2005) to account for bed geometry at low and high-interacting regimes,  
78 for Newtonian and non-Newtonian fluids.

79 Besides, another crucial feature in trickle-bed reactors is wetting efficiency as it determines  
80 the extent of catalyst utilization. Catalyst wetting is observed at two different scales: (i)  
81 external wetting at the particle scale and (ii) internal wetting at the pore scale. The former is  
82 the fractions of external catalyst surface covered by a liquid film, and the latter is the fraction  
83 of pores' surface covered by liquid. In trickle-bed reactors, complete internal wetting is usually  
84 assumed due to strong capillarity effects (Colombo et al. 1976). Thus, research works focused

85 on external wetting efficiency, henceforward referred to as wetting efficiency. Measurement  
86 techniques of wetting efficiency are either based on direct or indirect methods. The most  
87 widely used direct method is the dynamic tracer technique, which consists of injecting a dye  
88 tracer with the liquid phase into a trickle-bed-reactor, the wetted surface is then pigmented  
89 and measured to obtain the wetting efficiency. Although this approach is effective, it is usually  
90 applied in lab-scale reactors at low temperatures and pressures, operating at reduced  
91 superficial liquid velocities in order to keep the same liquid hourly space velocity as  
92 encountered in industrial-scale TBRs (Colombo et al. 1976; Burghardt et al. 1990; Al-Dahhan  
93 and Dudukovic 1995; van Houwelingen et al. 2006; Baussaron et al. 2007b; Baussaron et al.  
94 2007a; Julcour-Lebigue et al. 2009a). Concerning indirect methods, the chemical reaction  
95 method is the most used technique to measure implicitly the wetting efficiency. Unlike the  
96 direct methods, this method is not limited to low temperature and pressure conditions, it is  
97 applicable under industrial operating conditions (Schwartz et al. 1976; Sicardi et al. 1980; Al-  
98 Dahhan and Dudukovic 1995; Burghardt et al. 1995). However, a reactor model is needed to  
99 obtain wetting efficiencies through Residence Time Distribution (RTD), leading to a higher  
100 uncertainty.

101 Despite the fact that commercial reactors have an external wetting efficiency close to unity  
102 (Julcour-Lebigue et al. 2009a), many scale-up and hydrodynamic models are based on  
103 laboratory or pilot scale. Colombo et al. 1976 carried experimental tests in order to evaluate  
104 both external (at the pellet surface) and internal (inside the pellet's pores) effective wetting,  
105 the latter being found close to total due to capillary forces, whatever the wetting at the  
106 catalyst surface. Mills and Dudukovic (1981) reported that the increase of superficial liquid  
107 velocity increased wetting efficiency, achieving unity at high velocities. It was also the first

108 time a bounded function (guaranteeing  $0 < f < 1$ ) was proposed, using several dimensionless  
109 numbers.

110 More correlations were developed, many among them give dispersed results when  
111 analysing the same operating conditions. van Houwelingen et al. 2006 used the colorimetric  
112 method to study the effect that pre-wetting conditions had on particle wetting, which may be  
113 one of the reasons why the discrepancies in wetting efficiency values happen. In addition,  
114 many of the techniques employed didn't receive appropriate evaluations regarding their  
115 precision and thus dispersed results appear. (Julcour-Lebigue et al. 2009b) modelled wetting  
116 efficiency accounting for several relevant parameters, such as liquid viscosity, bed porosity,  
117 particle diameter and wall effects. However, the effect of gas velocity has not been  
118 investigated by the authors. Results showed that the effect of particle shape on wetting  
119 efficiency is almost negligible for the investigated shapes. Besides, an increase in bed porosity  
120 decreases external wetting efficiency, this is mainly due to reduction of particle-particle  
121 contact points.

122 In short, large discrepancies arise between some of the available correlations when  
123 applied in the same range of conditions, mainly due to the experimental conditions employed  
124 and to the use of complex techniques.

125 Further investigation is needed to predict parameters of interest and cover the TBR  
126 hydrodynamic grey areas, especially for large scale reactors under high pressure and  
127 temperature operating conditions. The purpose of the present work is to use a Computational  
128 Fluid Dynamics (CFD) approach to achieve this goal.

129 Research into modelling multiphase flows using CFD has gained pace in recent years,  
130 especially since the proof of promising predictions in complex conditions (Gunjal et al. 2005;  
131 Kuzeljevic and Dudukovic 2012; Gopal Manoharan and Buwa 2019; Jejurkar et al. 2020) are  
132 some CFD typical studies' examples. There are two main ways to approach multiphase flow in  
133 trickle-bed reactors as presented in the review of Wang et al. (2013): (i) Euler-Euler method  
134 and (ii) interface reconstruction method. The former method considers that the particle  
135 loading is represented by an effective porous medium. This approach results in lower  
136 calculation times and simulation numerical cost, at the extent of poor local description of  
137 particular non-ideal packing effects for instance. Solomenko et al. (2015) used the Euler-Euler  
138 method to simulate the liquid spreading in TBRs, and achieved a good agreement with volume-  
139 averaged experimental data in terms of liquid dispersion, even when the reactor is fed by a  
140 single-point liquid injection. This latter progress proved the relevance of CFD in predicting  
141 liquid distribution inside trickle-bed reactors. Later, Augier et al. (2017) coupled the  
142 hydrodynamic model of Solomenko et al. (2015) to a chemical reaction in order to study the  
143 effect of liquid maldistribution on reaction performances. Although this approach is effective  
144 in predicting phase distributions inside trickle-bed reactors, it has some disadvantages. The  
145 Euler-Euler approach does not enable to simulate flow at the particle scale, meaning that  
146 empirical or semi-empirical closure laws are needed to account for fluid/solid and fluid/fluid  
147 interactions. In response to this, the second approach - based on interface reconstruction - is  
148 increasingly used to simulate multiphase flows. Several numerical methods exist but the so-  
149 called Volume-Of-Fluid (VOF) approach is the most presented and used, as it is the most  
150 popular due to its simplicity and its possible use in complex geometries.

151 The VOF method allows tracking of explicitly defined fluid-fluid and fluid-solid interfaces,  
152 since it simulates flow over the actual physical geometry. Even though VOF method requires  
153 fine meshes, CFD studies have started using VOF method since it describes explicitly interfaces  
154 between the solid-liquid and liquid-gas phases, giving a more accurate description of the  
155 hydrodynamic phenomena. Augier et al. (2010) used the Volume-Of-Fluid method to  
156 investigate wetting and catalyst efficiencies in TBRs, the numerical domain consisted of three  
157 stacked spherical particles. A qualitative comparison between CFD and experimental flow  
158 snapshots shows a good prediction of the liquid distribution on the spheres. In addition, the  
159 predicted wetting efficiencies show similar tendencies as for experimental values. Haroun et  
160 al. (2012) used the VOF approach to improve the understanding of gas-liquid flow behaviour  
161 on structured packings, and its effect on mass transfer performances. The same approach was  
162 then used by Horgue et al. (2013) to predict the multiphase distribution on arrays of cylinders.  
163 A comparison between CFD phase distribution and experimental snapshots shows an accurate  
164 prediction of the flow topology. More recently, Deng et al. (2020) used the VOF approach to  
165 perform predictive simulations within packed loadings. The authors studied the effect of four  
166 different particle shapes as well as liquid properties effect on hydrodynamic parameters. The  
167 evolution of liquid holdup and wetting efficiency reported by the authors is in good agreement  
168 with previous observations in the literature. More recently Bouras et al. (2021) used VOF  
169 approach to simulate gas-liquid flow in structured multiphase reactors. A good agreement has  
170 been reported between experimental data and CFD simulations.

171 However, the VOF method has both advantages and limitations, it requires fine-enough  
172 meshes to avoid numerical diffusion at the fluid/fluid and fluid/solid interfaces, meaning that  
173 the required computational time might be very high. In addition, handling capillary flows with

174 VOF method is intricate as spurious velocities and fluxes emerge at the interfaces for high  
175 surface tension values. Despite all these limitations, the literature shows that VOF approach  
176 is reliable and efficient.

177       Nowadays, with the development of computational resources, reasonable computing  
178 times can be reached with more complex geometries. The VOF method can now be applied  
179 on larger volumes of trickle-bed reactor geometries, involving hundreds of solid particles, and  
180 generated using DEM simulation software as suggested by Boccardo et al. (2015). This article  
181 aims to answer two main interrogations: first, whether the VOF approach would predict  
182 pressure drop and trickling flow in representative volume of TBRs with a good accuracy and  
183 second, whether it can complete information obtained with direct experimental method to  
184 develop more precise wetting efficiency correlations.

185       To answer these questions, the first step is to build a realistic model of a 3D TBR, where  
186 hydrodynamic phenomena are simulated using CFD and compared with existing correlations.  
187 In this study, about 40 points were simulated, for different fluid systems, operating conditions  
188 and particle shapes. The simulations results have been compared to well validated models  
189 from the literature. Based on the simulation data and experimental data of Julcour-Lebigue  
190 et al. (2009a), a new correlation to predict an overall external wetting efficiency is proposed.

## 191 ***2. Numerical model***

192       This work uses Computational Fluid Dynamics in order to predict two-phase flow within  
193 trickle-bed reactors. A Volume-Of-fluid approach (Hirt and Nichols (1981)) is selected to  
194 accurately describe fluid-fluid and fluid-solid interfaces. The simulations are conducted using  
195 a commercial software ANSYS Fluent 19.2. In order to model the flow of immiscible fluids, the  
196 continuity equation (Eq. (1)) and the momentum equation (Eq. (2)) are solved simultaneously.

197 It is assumed that the fluids are Newtonian and incompressible, without phase change or heat  
 198 transfer. In addition, surface tension is assumed to be uniform at the fluids' interface.

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{u}) = 0 \quad (1)$$

$$\frac{\partial}{\partial t} (\rho \vec{u}) + \nabla \cdot (\vec{u} \vec{u}) = -\nabla p + \nabla (\mu (\nabla \vec{u} + \nabla \vec{u}^T)) + \rho \vec{g} + \vec{T}_\sigma \quad (2)$$

199 where  $\vec{u}$  is the velocity,  $p$  is the pressure,  $\mu$  and  $\rho$  are respectively the mixture viscosity and  
 200 density,  $\vec{g}$  is the gravitational acceleration and  $\vec{T}_\sigma$  is the surface tension force. The mixture  
 201 density and viscosity are calculated using the following equations (Equation (3)) and (4))

$$\rho = \sum_i \rho_i \alpha_i \quad (3)$$

$$\mu = \sum_i \mu_i \alpha_i \quad (4)$$

202 where  $\alpha_i$  is the volume fraction of the  $i$ -th phase. Interfaces' tracking is enabled through  
 203 resolution of the continuity equation of the volume fraction transport for one (or more)  
 204 phases. For the  $q^{th}$  phase, volume fraction transport is expressed as:

$$\frac{1}{\rho_q} \left[ \frac{\partial}{\partial t} (\rho_q \alpha_q) + \nabla \cdot (\rho_q \alpha_q \vec{u}) = S_{\alpha_q} \right] \quad (5)$$

205 where  $S_{\alpha_q}$  is the external mass source entering phase  $q$ . This equation is not solved for the  
 206 primary phase, its volume fraction is computed using the constraint in Equation (6):

$$\alpha_L + \alpha_G = 1 \quad (6)$$

207 Volume fraction  $\alpha_L$  is unity when only the liquid phase is present in the computational cell  
 208 and zero when only the gas phase is present in that cell. The in-between values refer to the  
 209 gas-liquid interface.

210 To account for capillarity effect, the Continuum Surface Force method (CSF) is employed.  
 211 This non-conservative method is implemented through a source term  $\vec{T}_\sigma$  in the momentum

212 equation. Proposed by Brackbill et al. (1992), the method assumes that surface tension is a  
213 continuous volume force. To further describe capillarity effects, a contact angle is specified at  
214 the catalyst's wall. Combined with surface normal, the contact angle allows to specify the  
215 interface's curvature, necessary to adjust the surface tension force  $\vec{T}_\sigma$ . Given the complexity  
216 of contact angle measurements in porous particles, a low contact angle value is usually  
217 employed (Augier et al. (2010)). This numerical choice is supported by experimental  
218 observations, since porous particles behave like a sponge and are commonly assumed to be  
219 fully wetted internally. For the sake of simplicity, a static contact angle is applied at the solid  
220 surface as it has been proved to give satisfying results by Augier et al. (2010). In this study a  
221 contact angle of  $1^\circ$  is specified at the catalyst particle surface.

222 This surface normal one cell away from the wall is given by Equation (7).

(7)

$$\vec{n} = \vec{n}_w \cos\theta_w + \vec{t}_w \sin\theta_w$$

223 where  $\theta_w$  is the contact angle at the wall, and  $\vec{n}_w$  and  $\vec{t}_w$  are the unit vectors normal and  
224 tangential to the wall respectively.

225 The simulations are conducted in transient conditions. Since trickle-bed reactors usually  
226 operate at low liquid Reynolds numbers, laminar flow conditions are considered with a  
227 pressure-based solver. The PISO scheme is employed to ensure pressure-velocity coupling,  
228 while PRESTO! is selected for pressure interpolation. The momentum equations are  
229 discretised using Second Order Upwind method. The resolution of two-phase flow throughout  
230 trickle-beds is achieved through the Volume-Of-Fluid approach, using the geo-reconstruct  
231 scheme. Finally, to guarantee numerical stability and convergence, a variable time step  
232 configuration is employed in order to maintain CFL condition under 1.

233 In order to simulate hydrodynamic performance within the considered loadings, boundary  
234 conditions shown in Figure 1 are considered. Regarding the multiphase flow, the gas and liquid  
235 enter the domain from the top, where their corresponding inlet velocities are specified. To  
236 describe the flow near the solid particles, a no-slip boundary condition is applied at the  
237 catalyst's surface. Since the numerical domains were extracted from larger loadings, the  
238 lateral boundaries are set to symmetry-type conditions. Finally, at the bottom, a regular  
239 pressure outlet condition is applied.

240 Packed bed loadings were generated in the open-source software Blender, as  
241 explained in the work of Boccoardo et al. (2015). Considering a particle shape, the packing  
242 generation is grounded on the Bullet Physics Library (BPL) available in Blender. This advanced  
243 physics simulation library solves the Newton's second law for a number of particles  $N$ , and is  
244 able to detect collisions between solid particles as well as their final position. According to  
245 Boccoardo et al. (2015), Blender is capable of handling complex particle shapes. Therefore,  
246 three particle shapes are considered in this work, namely spheres, trilobes and quadrilobes.

247 Spherical particles are loaded in a mono-dispersed configuration, meaning that all the spheres  
248 have equal diameters. In contrast, poly-dispersion is included in trilobes and quadrilobes  
249 loadings, with a mean particle length of 4.8mm and standard deviation of 1.8mm. The  
250 packings were generated within cylindrical containers of equal diameter ( $D=3.5\text{cm}$ ) and height  
251 ( $H=3\text{cm}$ ), their characteristics are summarized in Table 1. Spheres and trilobes loadings have  
252 similar void fractions, while the quadrilobes loading is characterised by a higher void fraction,  
253 as can be seen from Table 1. Since loadings were generated following the exact same  
254 procedure, high quadrilobe bed porosity is attributed to the particle's geometrical features.  
255 In contrast with spheres and trilobes, quadrilobes exhibit larger concavities.

256 In order to decrease computational cost, cuboid Representative Elementary Volumes (REV)  
257 are extracted from larger loadings in cylindrical containers. In order to obtain a surface-fitted  
258 mesh, the REV's are meshed using SnappyHexMesh following the recommendations of  
259 Boccardo et al. (2015), as shown in Figure 2.

260 The meshes were generated following a set of constraint on mesh quality metrics to avoid  
261 skewed and/or low orthogonal quality mesh cells. In addition, the mesh was refined near the  
262 catalyst surfaces to resolve sufficiently the contact points between particle shapes.  
263 Consequently, the contact points were represented by bridges to ensure all previous criteria  
264 are respected. Since VOF is sensitive to mesh quality, SnappyHexMesh was used to generate  
265 the different meshes. The artificial bridges between particles represent less than 1% of the  
266 total void volume.

267 Three different fluid systems were considered in this study: (i) heptane-nitrogen at  
268 standard conditions, (ii) a gas-liquid hydrocarbon mixture with properties commonly  
269 encountered in the middle distillate hydrocracking processes, and (iii) isohexadecane-nitrogen  
270 at standard conditions. The properties of the aforementioned systems are presented in Table  
271 2.

272 It is important to note that simulations were carried out in transient regime to prevent  
273 numerical instabilities or poor stabilization of transient phenomena. However, the results  
274 were analyzed after reaching steady state. The latter state is achieved after 3 second of flow  
275 time within the domain.

## 276 ***3. Numerical resolution***

### 277 ***3.1. Mesh convergence study***

278 As explained previously, the VOF approach requires a good quality mesh to provide accurate  
279 results. Therefore, a mesh independence study was performed on a representative volume of  
280 spheres in order to find the optimum mesh size. Four mesh densities are tested: (i) 1.5 Million,  
281 (ii) 3.4 Million, (iii) 4.3 Million and (iv) 11.2 Million cells. In order to capture the geometrical  
282 details, particularly at the solid-solid contact points, these meshes present a similar  
283 refinement level near the catalyst wall. The mesh independence study is conducted for  
284 constant liquid superficial velocity  $V_{S,L}$  and gas superficial velocity  $V_{S,G}$  conditions of:  $V_{S,L} =$   
285  $0.5\text{cm/s}$  and  $V_{S,G} = 5\text{cm/s}$ .

286 Figure 3 shows the level of refinement in each mesh and Table 3 summarises their  
287 corresponding resolutions and wetting efficiencies. Following the increase of mesh resolution,  
288 the predicted wetting efficiency values increase until reaching a plateau. In addition, almost  
289 tripling mesh density (from 4.3M to 11.2M) increases wetting efficiency by only 1.37%. It is  
290 interesting to note that simulation time increases from two days (4.3M) to fourteen days  
291 (11.2M) at constant CPU usage (128 cores).

292 Figure 3 illustrates contour plots of liquid volume fraction within the numerical domain  
293 for the considered meshes. The plots show clearly that the gas-liquid and liquid-solid  
294 interfaces are more distinguished for higher mesh resolutions. This means that numerical  
295 diffusion decreases with mesh resolution. Indeed, the coarser mesh (1.7 Million) exhibits  
296 considerable numerical diffusion, leading to a non-realistic representation of the gas-liquid  
297 interface. In fact, as the grid gets sharper, numerical thicknesses decrease, resulting in sharp  
298 fluid-fluid and fluid-solid interfaces.

299 In addition, mesh sharpness ensures a good definition of the boundary layer near the  
300 particles. As a result, the real particle loading geometry is better fitted, particularly at the

301 contact points between particles. The selected gas and liquid velocities lead to the most  
302 severe conditions that are very sensitive to mesh resolution. The lowest liquid velocity is the  
303 lowest liquid film thickness is. This case requires fine mesh resolution in the thin liquid film to  
304 describe properly hydrodynamic properties.

305 The 4.3 Million cell mesh gives similar results to those obtained with the 11.2 Million  
306 cell mesh, as shown in Table 3. In addition, both meshes allow a sharp description of the gas-  
307 liquid interface. However, when compared to the 4.3 Million cell mesh, the 11.2 Million cell  
308 mesh takes 6 to 7 times more time to reach convergence. Therefore, the 4.3 Million cell mesh  
309 is employed in the hereinafter simulations since it provides a trade off between results'  
310 precision and reasonable calculation times.

### 311 ***3.2. Inlet Effect on gas-liquid flow distribution and stabilization*** 312 ***within the calculation domain***

313 Liquid inlet effect on hydrodynamics is studied in order to identify the flow establishment zone  
314 within calculation domains. Indeed, since the liquid feed consists of 5 point sources, liquid  
315 distribution might require a certain bed depth to stabilise. Also called "calming bed depth",  
316 this stabilisation distance is achieved when liquid reaches a steady distribution. In order to  
317 check the distance needed for flow stabilisation, wetting efficiency is monitored in different  
318 regions of the numerical domains for HDT flow in several  $V_{S,L}$  and  $V_{S,G}$  conditions.

319 The numerical domain is divided into four sub-domains: (i) from 0 to 25% of the length (L), (ii)  
320 from 25%L to 50%L, (iii) from 50%L to 75%L and (iv) from 75%L to 100%L, given that inlet is  
321 located at  $z=0$  and outlet at  $z=L$ . Then, the wetted solid surface is calculated within each sub-  
322 domain as the integral of liquid volume fraction with respect to the solid surface. This study  
323 was conducted for all particle shapes at different operating conditions. However, results are

324 presented only for spherical shape since equivalent behaviours are found for all particle  
325 shapes.

326 Figure 4 presents wetting efficiency evolution obtained at different bed depths for spherical  
327 particles' loading. In the investigated  $V_{S,L}$  and  $V_{S,G}$  conditions, it can be observed that wetting  
328 efficiency reaches a plateau for bed depths beyond 50%L, indicating that the developed flow  
329 is reached. Indeed, wetting efficiency increases on average by 6% between (i) and (ii) sub-  
330 domains, then by 1.49% from (ii) to (iii) sub-domains, and finally by 0.03% from (iii) to (iv) sub-  
331 domains. Moreover, wetting efficiency follows a similar evolution regardless of gas and liquid  
332 superficial velocities. Due to complex phase interactions occurring near the inlet, and in order  
333 to achieve accurate and consistent results analysis, post-processing results presented  
334 hereinafter are computed within the developed flow domain only.

## 335 **4. Results and discussions**

### 336 **4.1. Pressure drop predictions**

337 A preliminary study was performed in order to validate pressure drop across the  
338 reactors. First, liquid downward flow was simulated in single-phase conditions for various  
339 Reynolds numbers at steady-state. These simulations were performed for loadings of spheres,  
340 trilobes and quadrilobes. Regarding boundary conditions, liquid superficial velocities ranging  
341 from 0.02mm/s to 30.75mm/s are investigated. The latter superficial velocities correspond to  
342  $0.05 < Re'_L < 100$ , where  $Re'_L$  is a modified Reynolds number defined as follows:

$$Re'_L = \frac{V_{S,L} d'_e \rho_L}{\mu_L (1 - \varepsilon_B)} \quad (8)$$

343  $d'_e$  is a modified equivalent diameter of the particles defined as the following:

$$d'_e = \Psi \cdot d_e = \Psi \cdot \frac{6V_p}{S_p} \quad (9)$$

344 Since Ergun law [Ergun 1952] was originally developed for spherical particles, a sphericity  
 345 factor is required to apply Ergun law [Ergun 1952] on non-spherical particles [Trahan et al.  
 346 2014]. Indeed, the sphericity factor  $\Psi$  given by equation (10) is a measurement of how close  
 347 a particle's shape is to a sphere.

$$\Psi = \frac{\pi^{\frac{1}{3}}(6V_p)^{\frac{2}{3}}}{S_p} \quad (10)$$

348 where  $V_p$  and  $S_p$  are respectively the particle's volume and surface area. This factor is  
 349 described as the ratio of (i) the surface area of a sphere with the same volume as the given  
 350 particle to (ii) the surface area of the non-spherical particle.

351 Pressure drop estimations in porous media are achieved through Ergun law [Ergun  
 352 1952]. It is possible as well to estimate pressure drop from CFD simulations. To do so, the  
 353 first step is to define two horizontal plane cuts within the developed-flow domain, at  
 354 different height locations ( $z_1$  and  $z_2$ ). Total pressure is then area-averaged within each  
 355 plane ( $P_1$  and  $P_2$ ), and pressure drop is obtained as  $\Delta P = \frac{P_1 - P_2}{z_1 - z_2}$ . A comparison between  
 356 CFD results and Ergun law [Ergun 1952] estimations is presented in Figure 5. The parity  
 357 plot highlights the accuracy of CFD in predicting single-phase pressure drop, since 95% of  
 358 results are within  $\pm 20\%$  and the majority is close to the identity line.

359 After checking the ability of CFD to handle pressure drop in single phase flow conditions,  
 360 the two-phase flow behaviour through three different loadings was predicted using the  
 361 Volume-Of-Fluid approach. Table 4 summarises the investigated gas and liquid superficial  
 362 velocities, as well as particle shapes. Thus, for each particle shape, 9 different ( $V_{S,L}, V_{S,G}$ )  
 363 couples were simulated for the hydrocarbon-hydrogen system.

364 Boyer et al. (2007) investigated pressure drop and liquid saturation in two-phase flow in  
365 the trickling regime and proposed predictive models for these two physical parameters. The  
366 authors validated these models over several experimental conditions and for different catalyst  
367 shapes. A two-phase pressure drop parity plot between CFD results and the model of Boyer  
368 et al. (2007) is presented in Figure 6.

369 The results show a very good agreement between CFD simulation and the model. Indeed,  
370 80% of the plotted points lie within the  $\pm 20\%$  deviation envelope, with an average relative  
371 error of 12.78. Pressure drop obtained with quadrilobes is low in comparison to spheres and  
372 trilobes as shown in Figure 6. This behaviour is expected due to the high void fraction of the  
373 quadrilobe particles' loading. In addition, it is observed that the most significant deviation  
374 between correlation and simulation results is obtained at the highest  $V_{S,G}$  values. The  
375 deviation could be attributed to a transition from laminar to turbulent flow, which is not  
376 accounted for in these CFD simulations.

## 377 **4.2. Gas-liquid flow behaviour in trickle bed reactors**

### 378 **4.2.1. Liquid saturation**

379 Similarly to two-phase pressure drop, liquid saturation is computed from the predicted  
380 liquid volume flowing through the particule loadings in established flow region. Figure 7 shows  
381 typical multiphase flow inside the investigated representative volumes; after reaching  
382 developed flow conditions. In the results presented hereinafter, the liquid saturation is  
383 calculated by the ratio of CFD liquid volume to bed void volume.

384 For both investigated fluid systems, i.e hydrotreatment and heptane-nitrogen systems,  
385 predicted liquid saturation values are compared to the values calculated by the correlation of

386 Boyer et al. (2007), as shown in Figure 8. Even though CFD results slightly over-estimate liquid  
387 saturation compared to Boyer et al. (2007) model's predicted value, the simulation results  
388 show an interesting agreement with an average relative deviation of 34%. It is interesting to  
389 note that the highest deviations are obtained for quadrilobes. It is also important to point out  
390 that the model of Boyer et al. (2007) has not been validated on this type of shape, which can  
391 explain the observed higher deviations. Moreover, it would be interesting to confront these  
392 CFD predictions to experimental data, not yet available.

393 From these results, it is observed that the higher the gas superficial velocity is, the lower  
394 the liquid saturation becomes. Such effect is a result of increased shear force at the gas-liquid  
395 interface, subsequently decreasing liquid film thickness through the particle loading. In  
396 addition, at constant gas superficial velocity, increasing liquid velocity results in increasing  
397 liquid saturation since liquid films become thicker as expected. Furthermore, quadrilobe  
398 shaped particles have particularly deep concavities leading to liquid accumulation (cf. Figure  
399 7). Therefore, higher liquid saturation values are obtained for quadrilobes. Besides, in  
400 comparison to spheres and trilobes, the void fraction was previously reported to be higher for  
401 the quadrilobe loading, promoting lower interstitial gas velocities within the particle bed, and  
402 thus higher liquid saturations.

#### 403 ***4.2.2. Liquid film thickness and flow regime investigation***

404 It is well known that the liquid film thickness has significant effect on mass transfer and  
405 thus is of high importance in trickle-bed reactors. Since CFD allows access to the local flow  
406 parameters, the CFD predicted average liquid film thicknesses are compared to the ones  
407 predicted by Nusselt's falling film theory estimations [Nusselt, 1916] assuming total catalyst  
408 wetting, as shown in Figure 9. Initially developed for liquid flow over infinite vertical planes,

409 Nusselt's theory describes the evolution of liquid film thickness with liquid superficial velocity.  
410 This theory is assumed to be a reasonable reference candidate to describe liquid flow over  
411 complex surfaces, such as particle loadings. Commonly, in steady-state conditions, the  
412 average liquid film thickness is defined as the ratio of liquid volume to the wetted catalyst  
413 surface. To avoid inlet effects, liquid film thickness is computed within the developed flow  
414 domain.

415 Results plotted in Figure 9 highlight the effect of gas and liquid superficial velocities on  
416 liquid film thickness. Indeed, the results show that increasing liquid superficial velocities leads  
417 to thicker films, while the increase of gas flow rate results in the opposite effect. These  
418 conclusions are consistent with previous observations on liquid saturation. As the gas velocity  
419 increases, the gas-liquid shear interactions are enhanced, resulting in liquid entrainment and  
420 liquid film thickness reduction. Besides, for identical superficial velocities, liquid film  
421 thicknesses are on average higher for quadrilobes than the rest of the shapes. As was  
422 discussed previously, quadrilobe's geometrical characteristics (concavities) as well as lower  
423 interstitial gas velocities (high bed porosity) tend to promote an increase in film thickness.

424 Furthermore, results show that CFD and Nusselt's model (Nusselt 1916) predict a  
425 predominant effect of liquid superficial velocity on liquid film thickness. Thus, the model can  
426 be used to obtain an order of magnitude of the mean liquid film thickness, since mean relative  
427 deviations of 14.7%, 18.1% and 20.6% are achieved respectively for spheres, trilobes and  
428 quadrilobes. Nevertheless, the effect of  $V_{S,G}$  is not captured by the model.

### 429 ***4.2.3. Wetting efficiency predictions***

430 Wetting efficiency is defined as the ratio of predicted wetted solid area to total solid area. For  
431 the sake of consistency, wetting efficiencies are computed in the developed flow domain,  
432 beyond the calming depth. The results are compared to the correlation of Julcour-Lebigue et  
433 al. (2009a), given by the following equation:

$$f = 1 - \exp[-1.986Fr_L^{0.139}Mo_L^{0.0195}\varepsilon_B^{-1.55}] \quad (11)$$

434 where  $\varepsilon_B$  is the bed porosity,  $Fr_L$  and  $Mo_L$  are respectively the liquid Froude and Morton  
435 dimensionless numbers, which accurately describe both liquid velocity and physical properties  
436 effects.

437 Figure 10 shows a comparison between wetting efficiencies obtained by CFD and the  
438 correlation of Julcour-Lebigue et al. (2009a). Several observations arise from the analysis of  
439 this parity plot. For spheres and trilobes, predicted wetting efficiency values are in a good  
440 agreement with the correlation of Julcour-Lebigue et al. (2009a) at low gas superficial  
441 velocities. However, a significant deviation is observed for quadrilobe particles as the offset  
442 between CFD and correlation values is much higher than 20%. Although Julcour-Lebigue et al.  
443 (2009a) developed a wetting efficiency correlation for different particle shapes, their  
444 investigation did not include quadrilobe shaped particles. In addition, the authors performed  
445 experiments in the absence of gas flow, thus equation **(11)** does not account for gas superficial  
446 velocity  $V_{S,G}$  effect. For these reasons, high deviations between CFD and correlation are  
447 reported in Figure 10.

448 It is interesting to note that CFD predictions show a negative impact of gas velocity  $V_{S,G}$  on  
449 wetting efficiency. This is attributed to liquid disruption resulting in low liquid adherence to  
450 the solid surface. Wetting efficiency decreases with gas superficial velocities has also been  
451 observed in other CFD simulations, more recently on the work of Deng et al. (2020). The

452 authors argued that these decreases are mainly due to higher gas-liquid interactions. Similarly  
 453 Herskowitz and Mosseri (1983) observed a global negative impact of  $V_{S,G}$  on the global rate of  
 454 a catalytic reaction. The proposed explanation was the increase of dry catalyst area with gas  
 455 velocity, thus reducing the apparent effectiveness of the catalyst. This has been confirmed by  
 456 Burghardt et al. (1990), who employed a mathematical description of the dynamic tracer  
 457 method in order to review literature results. The authors reached the same observations.

458 In this context, a new correlation is developed on the basis of CFD simulations performed in  
 459 this work and former experimental data of , Julcour-Lebigue et al. (2009a) to include the effect  
 460 of catalyst shape and gas superficial velocity

#### 461 ***4.2.4. Development of a new wetting efficiency correlation***

462 In their work, Julcour-Lebigue et al. (2009a) correlated wetting efficiency to three parameters,  
 463 namely bed porosity  $\varepsilon_B$ , liquid Morton number  $Mo_L$  and Froude number  $Fr_L$ . In this section,  
 464 an improvement of the aforementioned correlation is proposed in order to include gas  
 465 velocity and particle shape effects. In addition to predictions of hydrotreatment system and  
 466 heptane-nitrogen, isohexadecane-nitrogen system was investigated by CFD in the conditions  
 467 presented in Table 4. In an effort to broaden physico-chemical properties, isohexadecane-  
 468 nitrogen system was selected for its high liquid viscosity and surface tension.

469 Analysis of simulation results unveiled important physical scales to account for in the  
 470 new correlation: (i) gas superficial velocity  $V_{S,G}$ , (ii) gas-phase properties ( $\rho_G, \mu_G$ ) and (iii)  
 471 particle characteristics ( $d_p, a_s$ ). Therefore, the proposed general formulation is given by:

$$f = 1 - \exp \left[ -A \times Fr_L^{0.139} Mo_L^{0.0195} \varepsilon_B^{-1.55} S^{C_0} \left( 1 - C_1 \prod_{i=2}^n N_i^{C_i} \right) \right] \quad (12)$$

472 where  $C_0, C_i, \dots, C_n$  are fitted constants,  $N_i$  are different gas-phase dimensionless groups,  $A$  is  
473 a multiplication factor and  $S$  is a shape factor.

474 Gas-phase dimensionless groups  $N_i$  are chosen from the following dimensionless numbers:  
475  $Re_G, Fr_G, Ga_G$ . Mathematical optimisation is conducted to determine the best group  
476 combination and fit the new correlation. The Sum of Squared Errors (SSE) between correlation  
477 and CFD/experimental values is minimised using a Generalised Reduced Gradient (GRG)  
478 algorithm. Table 5 summarises the obtained standard deviations with different group  
479 combinations. The minimum standard deviation was reached in Case 2 and Cases 4-7.  
480 Therefore, for the sake of simplicity, the gas Froude number is selected to include gas phase  
481 properties effect on wetting efficiency. It is important to point out that for liquid flow at  $V_{S,G} =$   
482  $0$ ,  $Fr_G$  is equal to zero and the correlation resumes to the model of Julcour-Lebigue et al.  
483 (2009a). Even though gas phase properties effect on the gas Froude number was not  
484 investigated in the scope of this work, the developed CFD model could be employed in a future  
485 work to establish this effect.

486 First, particle shape effect is described using a constant shape factor  $\phi$ , where  $\phi$  depends  
487 solely on the particle shape and is equal to unity for spherical particles. The following  
488 correlation is then obtained:

$$f = 1 - \exp\left[-1.986Fr_L^{0.139}Mo_L^{0.0195}\epsilon_B^{-1.55}\phi(1 - 0.437Fr_G^{0.367})\right] \quad (13)$$

489 Table 6 shows the various constant shape factors for each shape. As one can see, the equation  
490 is similar to the original one developed by Julcour-Lebigue et al. (2009). The correlation  
491 presented in equation (13) has not been evaluated for particle shapes other than spheres,

492 trilobes and quadrilobes. Therefore, it cannot be used to estimate the wetting efficiency for  
 493 any particle shape.

494 In order to expand the correlation of Julcour-Lebigue et al. (2009a) to different particle  
 495 shapes and sizes, a different shape factor  $S$  is introduced as shown in equation (14). First, the  
 496 shape factor  $a_S d_p / \varepsilon_B^2$  used by Mills and Dudukovic (1981) was tested, but a good wetting  
 497 efficiency estimation could not be reached. Nevertheless, a good fit was achieved using the  
 498 shape factor proposed by Ellman et al. (1990) :

$$S = \frac{a_S d_h}{1 - \varepsilon_B} \quad (14)$$

499 This factor accounts for particle's shape and size through the hydraulic diameter  $d_h$ . Whereas  
 500 bed characteristics are included through specific area of the bed  $a_S$  and its porosity  $\varepsilon_B$ .  
 501 Considering the aforementioned shape factor, the wetting efficiency correlation is expressed  
 502 as the following:

$$f_{corr} = 1 - \exp \left[ -0.649 Fr_L^{0.139} Mo_L^{0.0195} \varepsilon_B^{-1.55} \left( \frac{d_h a_S}{1 - \varepsilon_B} \right)^{1.147} (1 - 0.436 Fr_G^{0.371}) \right] \quad (15)$$

503 Ellman et al. (1990) observed that the shape factor proposed in equation (14) could be  
 504 further simplified since:

$$\frac{a_S d_h}{1 - \varepsilon_B} \propto const \times \frac{\varepsilon_B^3}{(1 - \varepsilon_B)^2} \quad (16)$$

505 Due to the proportionality observed in equation (16), the correlation expression given by  
 506 equation (15) can be further simplified. Another correlation expression, accounting for both  
 507 bed void fraction and particle shape, is presented in the following equation:

$$f = 1 - \exp \left[ -4.065 Fr_L^{0.139} Mo_L^{0.0195} \varepsilon^{-1.55} \left( \frac{\varepsilon_B^3}{(1 - \varepsilon_B)^2} \right)^{0.376} (1 - 0.434 Fr_G^{0.376}) \right] \quad (17)$$

508 The standard deviations for each correlation with different particle shape factors are  
509 shown in Table 7. It can be seen that very good agreement is achieved by either expression,  
510 since deviation is under 10%.

511 The same standard deviation value is reached using equation (15) and equation (17), owing to  
512 the proportionality observed between shape factors used in both correlation expressions.  
513 Wetting efficiency is estimated using equation (15), and compared to experimental and CFD  
514 results for over 110 data points. Figure 11 presents a parity diagram of correlation estimations  
515 against CFD and experimental results.

516 It is possible to observe from Figure 11 that all multiphase CFD results and almost all  
517 experimental results ( $V_{S,G} = 0$ ) are well fitted with the new developed correlation, even for  
518 more irregular particle shapes. It should be noted that higher deviations are observed for  
519 liquids -such as water and ethanol-. A corrective term was developed by Julcour-Lebigue et al.  
520 (2009a) which reasonably accounts for the contact angle effect through a critical liquid phase  
521 Froude number.

522 Without any doubt, Table 7 and Figure 11 confirm the good agreement between simulated  
523 and experimental values, highlighting the relevance of adding gas velocity and particle shape  
524 effects to the wetting efficiency correlation. Validity ranges of the wetting correlation are  
525 summarised in

526 Table 8, excluding water and ethanol.

## 527 **5. Conclusion**

528 In this work, Computational Fluid Dynamics were employed to investigate the  
529 influence of catalyst shape, fluids properties and gas-liquid flow operating conditions on  
530 pressure drop, liquid saturation, liquid flow behaviour, wetting efficiency in trickle-bed  
531 reactors.

532 A numerical model grounded on the Volume-Of-Fluid approach was used. The  
533 multiphase CFD results were compared to validated and well-established correlations from  
534 literature for pressure drop, liquid saturation, liquid film thickness and wetting efficiency. A  
535 good agreement was achieved between CFD results and existing correlations ((Boyer et al.  
536 2007) and (Julcour-Lebigue et al. 2009a)). Therefore, these results validated the numerical  
537 model and showed that CFD can be a powerful tool to predict complex flow patterns in trickle  
538 bed reactors. In addition, CFD allows access to local phenomena and spatial distributions of  
539 transport parameters within the loadings, such as liquid film thicknesses.

540 The CFD model is then employed to investigate effects of (i) gas and liquid velocity, (ii)  
541 gas and liquid properties and (iii) catalyst shape on flow behaviour and wetting efficiency. It is  
542 found that increasing gas velocity leads to the decrease of wetting efficiency, with stronger  
543 effects noticed at higher gas densities. This was already observed in previous studies (such as  
544 the study developed by Al-Dahhan and Dudukovic (1995)). Moreover, particle geometry effect  
545 on hydrodynamic parameters was found greater for irregular shapes (quadrilobes). Overall  
546 results analysis revealed the significant influence of gas superficial velocity and particle shape  
547 on hydrodynamic parameters, particularly wetting efficiency. Therefore, an expansion of the  
548 wetting efficiency correlation of Julcour-Lebigue et al. (2009a) is proposed. In order to account  
549 for gas velocity and particle shape effects, two additional dimensionless numbers are added  
550 into the initial correlation. The gas velocity effect is included through gas Froude number  $Fr_G$ ,

551 while particle shape effect is accounted for through two shape factor expressions, depending  
552 on available characteristics.

553 The new correlation, developed in this study, can predict wetting efficiency with good  
554 accuracy (less than 10% deviation) for CFD experiments and already published experimental  
555 results of Julcour-Lebigue et al. (2009a). The correlation is valid for a wide range of physico-  
556 chemical conditions, at high and low pressures, involving highly wetting liquid-solid systems  
557 as hydrocarbon and alumina. Further investigations may be required to extend the models to  
558 much less wetting liquid systems like water and ethanol.

559 In addition, this work shows that CFD VOF approach is an effective tool for the prediction of  
560 pressure drop, wetting efficiency and trickling flow in representative volume of TBRs, with a  
561 good accuracy and allow to develop more precise wetting efficiency correlations. A such work  
562 methodology presented in this work could be used for the innovation and optimisation of  
563 catalyst shape particles.

564

## 565 **6. Publication bibliography**

566 Al-Dahhan, M. H.; Dudukovic, M. P. (1995): Catalyst Wetting Efficiency in Trickle-Bed Reactors  
567 at High Pressure. In *Chemical Engineering Science* 50, pp. 2377–2389.

568 Augier, F.; Koudil, A.; Royon-Lebeaud, A.; Muszynski, L.; Yanouri, Q. (2010): Numerical  
569 approach to predict wetting and catalyst efficiencies inside trickle bed reactors. In *Chemical*  
570 *Engineering Science* 65 (1), pp. 255–260. DOI: 10.1016/j.ces.2009.06.027.

571 Augier, Frédéric; Fourati, Manel; Haroun, Yacine (2017): Characterization and modelling of a  
572 maldistributed Trickle Bed Reactor. In *Can. J. Chem. Eng.* 95 (2), pp. 222–230. DOI:  
573 10.1002/cjce.22618.

574 Bansal, A.; WANCHOO, R. K.; SHARMA, S. K. (2005): Flow Regime Transition in a Trickle Bed  
575 Reactor. In *Chemical Engineering Communications* 192 (8), pp. 1046–1066. DOI:  
576 10.1080/009864490522597.

577 Baussaron, L.; Julcour-Lebigue, C.; Wilhelm, A. M.; Boyer, C.; Delmas, H. (2007a): Partial  
578 Wetting in Trickle Bed Reactors: Measurement Techniques and Global Wetting Efficiency. In  
579 *Ind. Eng. Chem. Res.* 46 (25), pp. 8397–8405.

580 Baussaron, L.; Julcour-Lebigue, C.; Wilhelm, A. M.; Delmas, H.; Boyer, C. (2007b): Wetting  
581 topology in trickle bed reactors. In *AIChE J.* 53 (7), pp. 1850–1860.

582 Boccardo, G.; Augier, F.; Haroun, Y.; Ferré, D.; Marchisio, D. L. (2015): Validation of a novel  
583 open-source work-flow for the simulation of packed-bed reactors. In *Chemical Engineering*  
584 *Journal* 279, pp. 809–820. DOI: 10.1016/j.cej.2015.05.032.

585 Bouras, H.; Haroun, Y.; Philippe, R.; Augier, F.; Fongarland, P. (2021): CFD modeling of mass  
586 transfer in Gas-Liquid-Solid catalytic reactors. In *Chemical Engineering Science* 233.

587 Boyer, C.; Volpi, C.; Ferschneider, G. (2007): Hydrodynamics of trickle bed reactors at high  
588 pressure: Two-phase flow model for pressure drop and liquid holdup, formulation and  
589 experimental validation. In *Chemical Engineering Science* 62 (24), pp. 7026–7032. DOI:  
590 10.1016/j.ces.2007.08.036.

591 Brackbill, J. U.; Kothe, D. B.; Zemach, C. (1992): A Continuum Method for Modeling Surface  
592 Tension. In *Journal of Computational Physics* 100 (335-354).

593 Burghardt, A.; Bartelmus, G.; Jaroszynski, M.; Kolodziej, A. (1995): Hydrodynamics and Mass  
594 Transfer in a Three-Phase Fixed-Bed Reactor with Cocurrent Gas-Liquid Downflow. In *The*  
595 *Chemical Engineering Journal* 58, pp. 83–99.

596 Burghardt, A.; Kolodziej, A. S.; Jaroszynski, M. (1990): Experimental Studies of Liquid-Solid  
597 Wetting Efficiency in Trickle-Bed Cocurrent Reactors. In *Chemical Engineering and Processing*  
598 28, pp. 35–49.

599 Colombo, A. J.; Baldi, G.; Sicardi, S. (1976): Solid-liquid contacting effectiveness in trickle bed  
600 reactors. In *Chemical Engineering Science* 31, pp. 1101–1108.

601 Deng, H.; Guo, B.; Dong, H.; Liu, C.; Geng, Z. (2020): Computational Investigation of Liquid  
602 Holdup and Wetting Efficiency Inside Trickle Bed Reactors with Different Catalyst Particle  
603 Shapes. In *Applied Sciences* 10 (4), p. 1436.

604 Ellman, M. J.; Midoux, N.; Wild, G.; Laurent, A.; Charpentier, J. C. (1990): A New, Improved  
605 Liquid Hold-up Correlation for Trickle-Bed Reactors. In *Chemical Engineering Science* 45,  
606 pp. 1677–1684.

607 Ergun, S. (1952). Fluid flow through packed columns. In *Chemical Engineering Progress* 48, pp.  
608 89–94.

609 Gopal Manoharan, Karthik; Buwa, Vivek V. (2019): Structure-Resolved CFD Simulations of  
610 Different Catalytic Structures in a Packed Bed. In *Ind. Eng. Chem. Res.* 58 (49), pp. 22363–  
611 22375. DOI: 10.1021/acs.iecr.9b03537.

612 Gunjal, Prashant R.; Kashid, Madhavanand N.; Ranade, Vivek V.; Chaudhari, Raghunath V.  
613 (2005): Hydrodynamics of Trickle-Bed Reactors: Experiments and CFD Modeling. In *Ind. Eng.*  
614 *Chem. Res.* 44 (16), pp. 6278–6294. DOI: 10.1021/ie0491037.

615 Haroun, Y.; Raynal, L.; Legendre, D. (2012): Mass transfer and liquid hold-up determination in  
616 structured packing by CFD. In *Chemical Engineering Science* 75, pp. 342–348. DOI:  
617 10.1016/j.ces.2012.03.011.

618 Herskowitz, M.; Mosseri, S. (1983): Global rates of reaction in trickle-bed reactors: effects of  
619 gas and liquid flow rates. In *Industrial & Engineering Chemistry Fundamentals* 22, pp. 4–6.

620 Hirt, C. W.; Nichols, B. D. (1981): Volume Of Fluid (VOF) Method for the Dynamics of Free  
621 Boundaries. In *Journal of Computational Physics* 39, pp. 201–225.

622 Holub, R. A.; Dudukovic, M. P.; Ramachandran, P. A. (1992): A phenomenological model for  
623 pressure drop, liquid holdup, and flow regime transition in gas-liquid trickle flow. In *Chemical*  
624 *Engineering Science* 47, pp. 2343–2348.

625 Horgue, Pierre; Augier, Frédéric; Duru, Paul; Prat, Marc; Quintard, Michel (2013):  
626 Experimental and numerical study of two-phase flows in arrays of cylinders. In *Chemical*  
627 *Engineering Science* 102, pp. 335–345. DOI: 10.1016/j.ces.2013.08.031.

628 Jejurkar, Swarup Y.; Khanna, Ashok; Verma, Nishith (2020): Maldistribution Effects in an  
629 Industrial-Scale Trickle Bed Reactor. In *Ind. Eng. Chem. Res.* 59 (16), pp. 7405–7415. DOI:  
630 10.1021/acs.iecr.0c00115.

631 Julcour-Lebigue, C.; Augier, F.; Maffre, H.; Wilhelm, A. M.; Delmas, H. (2009a): Measurements  
632 and Modeling of Wetting Efficiency in Trickle-Bed Reactors: Liquid Viscosity and Bed Packing  
633 Effects. In *Ind. Eng. Chem. Res.* 48 (14), pp. 6811–6819.

634 Julcour-Lebigue, Carine; Augier, Frédéric; Maffre, Harold; Wilhelm, Anne-Marie; Delmas,  
635 Henri (2009b): Measurements and Modeling of Wetting Efficiency in Trickle-Bed Reactors.  
636 Liquid Viscosity and Bed Packing Effects. In *Ind. Eng. Chem. Res.* 48 (14), pp. 6811–6819. DOI:  
637 10.1021/ie9002443.

638 Kuzeljevic, Zeljko V.; Dudukovic, Milorad P. (2012): Computational Modeling of Trickle Bed  
639 Reactors. In *Ind. Eng. Chem. Res.* 51 (4), pp. 1663–1671. DOI: 10.1021/ie2007449.

640 Larachi, F.; Laurent, A.; Midoux, N.; Wild, G. (1991): Experimental Study of a Trickle-Bed  
641 Reactor Operating at High Pressure: Two-Phase Pressure Drop and Liquid Saturation. In  
642 *Chemical Engineering Science* 46, pp. 1233–1246.

643 Larkins, R. P.; White, R. R.; Jeffrey, D. W. (1961): Two-Phase Concurrent Flow in Packed Beds.  
644 In *AIChE J.* 7, pp. 231–239.

645 Lockhart, R. W.; Martinelli, R. C. (1949): Proposed Correlation of Data for Isothermal Two-  
646 Phase, Two-Component Flow in Piped. In *Chemical Engineering Progress* 45, pp. 39–48.

647 Midoux, N.; Favier, M.; Charpentier, J. C. (1976): Flow Pattern, Pressure Loss and Liquid Holdup  
648 Data in Gas-Liquid Downflow Packed Beds with Foaming and Nonfoaming Hydrocarbons. In  
649 *Journal of Chemical Engineering of Japan* 9, pp. 350–356.

650 Mills, P. L.; Dudukovic, M. P. (1981): Evaluation of Liquid-Solid Contacting in Trickle-Bed  
651 Reactors by Tracer Methods. In *AIChE J.* 27, 893-904.

652 Nishizawa, A.; Kitano, T.; Ikenaga, N.; Miyake, T.; Suzuki, T. (2014): Use of Trickle Bed Reactor  
653 for Fischer-Tropsch Reaction over Co-Mn/Oxidized Diamond Catalyst. In *Journal of the Japan*  
654 *Petroleum Institute* 57, pp. 109–117.

655 Nusselt, W. (1916): Die Oberflächenkondensation des Wasserdampfes. In *Z. Vereines Deutsch.*  
656 *Ing.* 60, 541-546,569-575.

657 Rao, V. G.; Ananth, M. S.; Varma, Y.B.G. (1983): Hydrodynamics of Two-Phase Cocurrent  
658 Downflow through Packed Beds. In *AIChE J.* 29, pp. 467–483.

659 Ratman, G.S.V.; Ananth, M. S.; Varma, Y.B.G. (1993): A Model for Pressure Drop in Gas-Liquid  
660 Cocurrent Downflow through Packed Beds. In *Chemical Engineering Journal* 51, pp. 19–28.

661 Sai, P.S.T.; Varma, Y.B.G. (1987): Pressure Drop in Gas-Liquid Downflow through Packed Beds.  
662 In *AIChE J.* 33, pp. 2027–2036.

663 Sato, Y.; Hirose, T.; Takahashi, F.; Toda, M. (1973): Pressure Loss and Liquid Holdup in Packed  
664 Bed Reactor with Cocurrent Gas-Liquid Downflow. In *Journal of Chemical Engineering of Japan*  
665 6, pp. 147–152.

666 Schwartz, J. G.; Weger, E.; Dudukovic, M. P. (1976): A New Tracer Method for Determination  
667 of Liquid-Solid Contacting Efficiency in Trickle-Bed Reactors. In *AIChE J.* 22 (5), pp. 894–904.

668 Sicardi, S.; Baldi, G.; Gianetto, A.; Specchia, V. (1980): Catalyst Areas Wetted by Flowing and  
669 Semistagnant Liquid in Trickle-Bed Reactors. In *Chemical Engineering Science* 35, pp. 67–73.

670 Solomenko, Z.; Haroun, Y.; Fourati, M.; Larachi, F.; Boyer, C.; Augier, F. (2015): Liquid spreading  
671 in trickle-bed reactors: Experiments and numerical simulations using Eulerian–Eulerian two-  
672 fluid approach. In *Chemical Engineering Science* 126, pp. 698–710. DOI:  
673 10.1016/j.ces.2015.01.013.

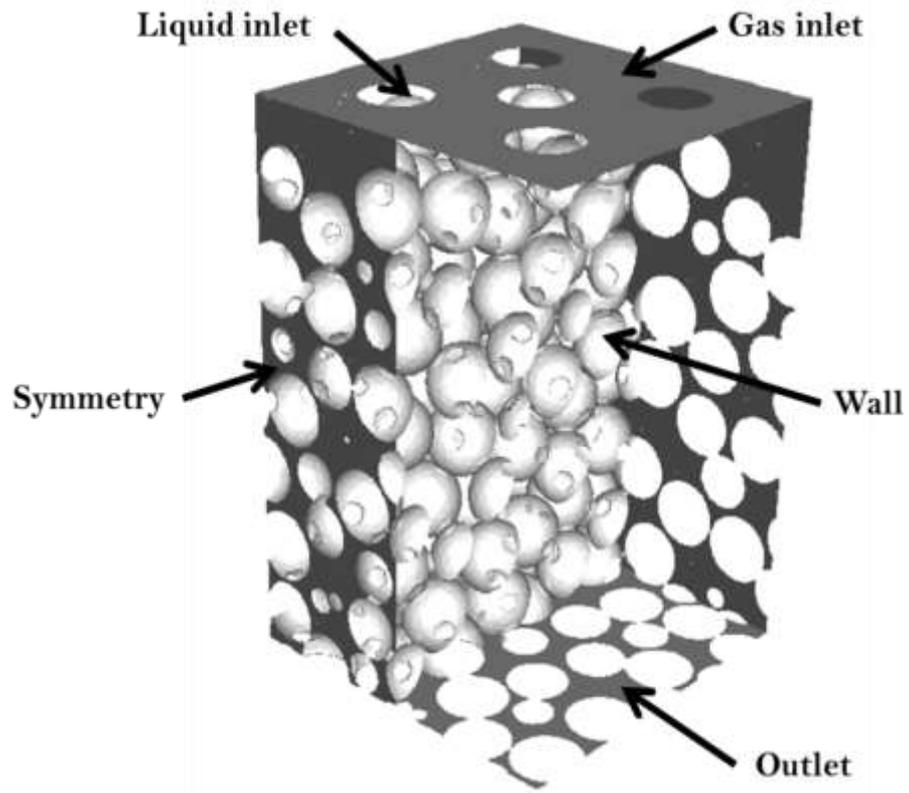
674 Specchia, V.; Baldi, G. (1977): Pressure Drop and Liquid Holdup for Two-Phase Concurrent Flow  
675 in Packed Beds. In *Chemical Engineering Science* 32, pp. 515–523.

676 Tosun, G. (1984): A Study of Cocurrent Downflow of Nonfoaming Gas-Liquid Systems in a  
677 Packed Bed. 2. Pressure Drop: Search for a Correlation. In *Industrial & Engineering Chemistry*  
678 *Process Design and Development* 23, pp. 35–39.

679 van Houwelingen, A. J.; Sandrock, C.; Nicol, W. (2006): Particle wetting distribution in trickle-  
680 bed reactors. In *AIChE J.* 52 (10), pp. 3532–3542. DOI: 10.1002/aic.10961.

681 Wang, Yining; Chen, Jinwen; Larachi, Faical (2013): Modelling and simulation of trickle-bed  
682 reactors using computational fluid dynamics: A state-of-the-art review. In *Can. J. Chem. Eng.*  
683 91 (1), pp. 136–180. DOI: 10.1002/cjce.20702.

684

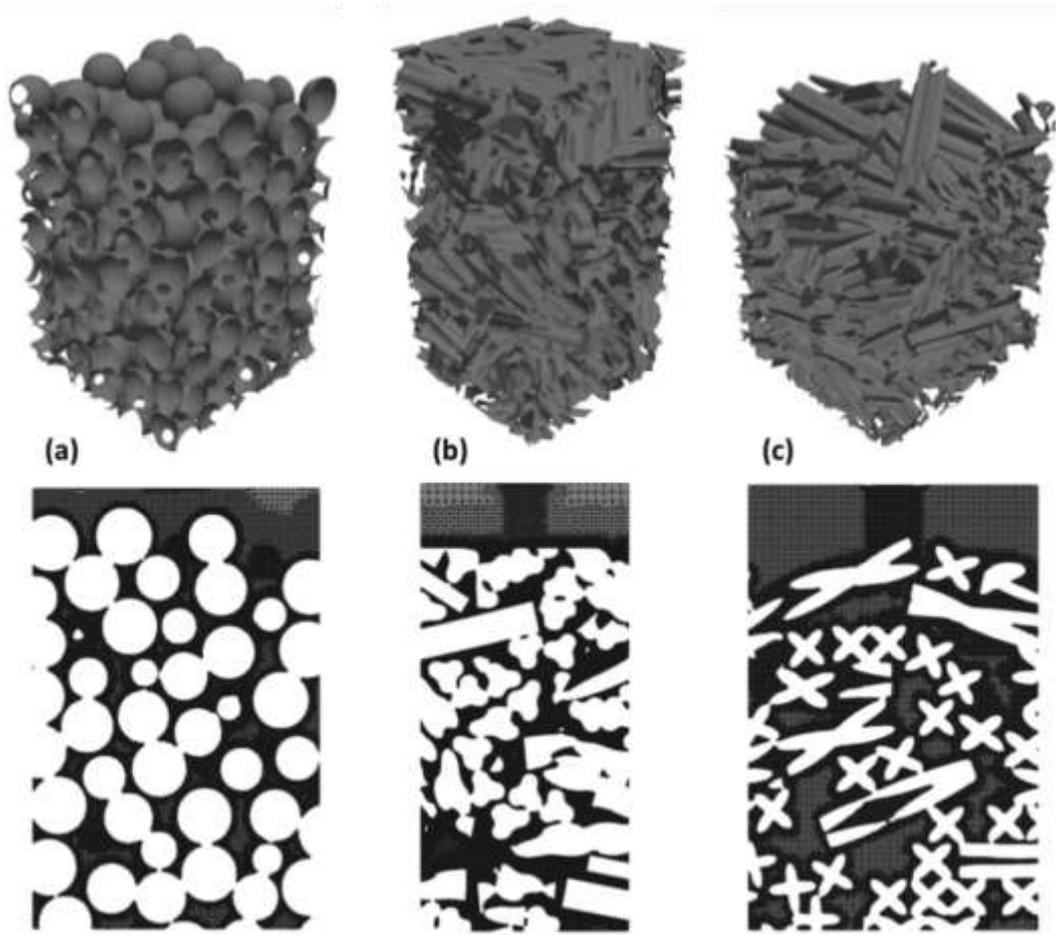


686

687

688

Figure 1: Numerical domain and boundary conditions description for the two-phase flow predictions. The figure shows a Representative Elementary Volume (REV) of  $1.0 \times 1.0 \times 1.5 \text{ cm}^3$  extracted from a spherical particles' loading.



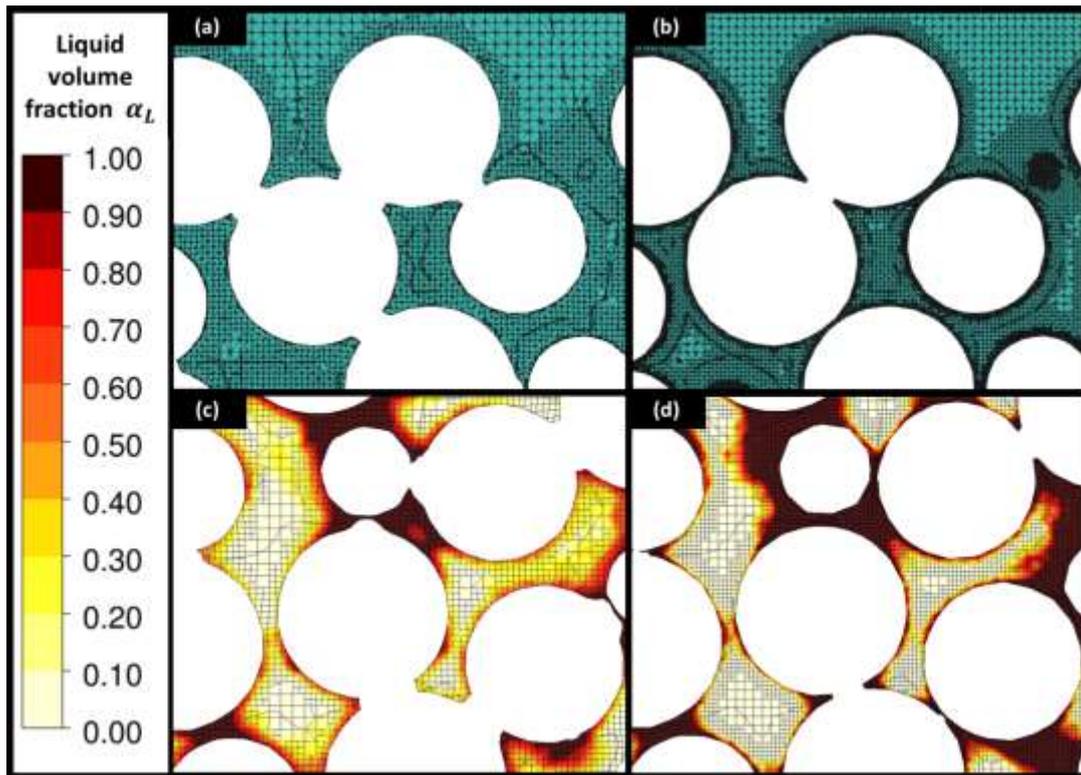
689

690

691

692

Figure 2: The numerical domains (upper row) and slices of their respective meshes (a) spheres ( $1.0 \times 1.0 \times 1.5 \text{ cm}^3$ ,  $d_e = 2.00 \text{ mm}$ ,  $\varepsilon_B = 37.2\%$ ) (b) trilobes ( $1.0 \times 1.0 \times 2.1 \text{ cm}^3$ ,  $d_e = 1.80 \text{ mm}$ ,  $\varepsilon_B = 37.7\%$ ) and (c) quadrilobes ( $1.0 \times 1.0 \times 1.5 \text{ cm}^3$ ,  $d_e = 1.95 \text{ mm}$ ,  $\varepsilon_B = 62.8\%$ ).



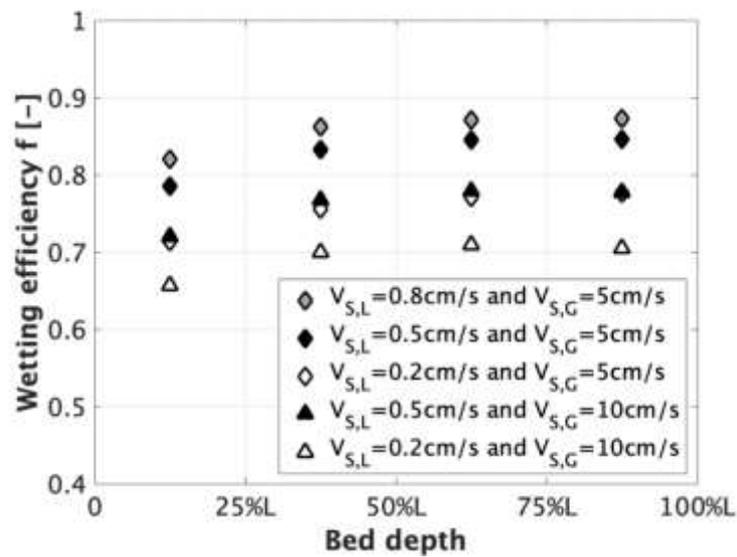
693

694

Figure 3: Minimum and maximum mesh densities of the spherical loading: (a) 1.7Million, (b) 11.2Million cell meshes

695

and Liquid fraction contour plot for (c) 1.7Million (d) 11.2Million cell meshes.



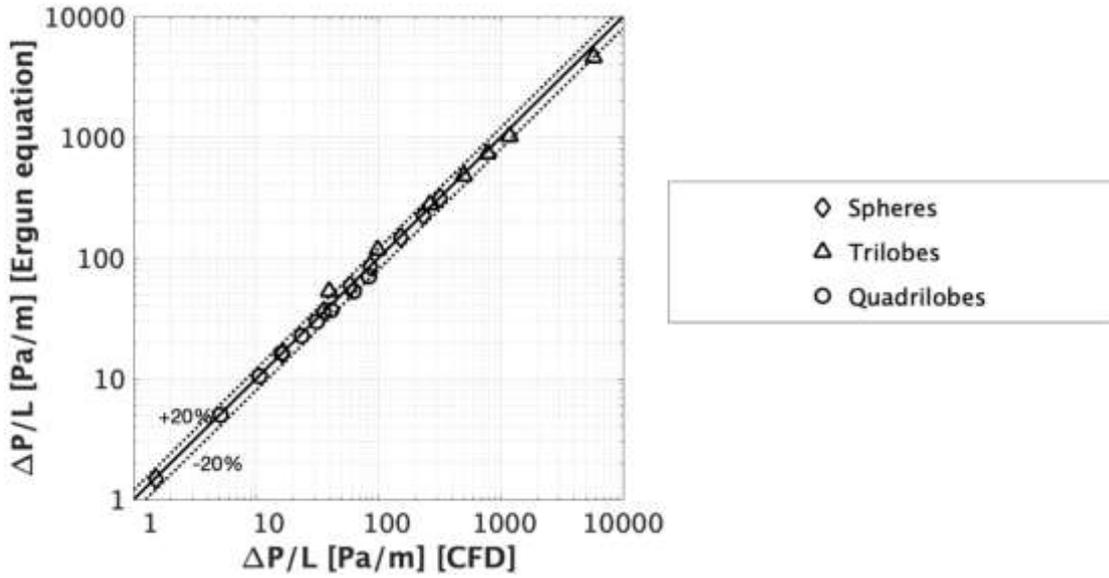
696

697

Figure 4: Wetting efficiency evolution within spherical particles' packing for HDT flow in different gas and liquid

698

superficial velocities conditions.

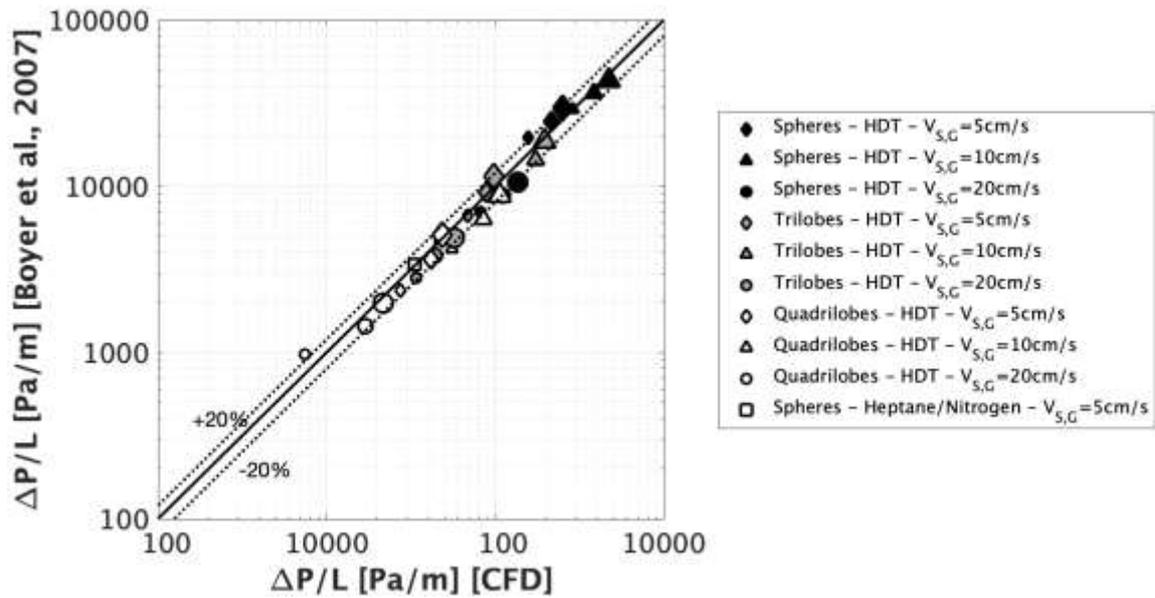


699

700

Figure 5: Parity diagram of single-phase pressure drop comparison between Ergun and CFD. Sphericity factors are respectively  $\Psi = 1$ ,  $\Psi = 0.81$  and  $\Psi = 0.42$  for spheres, trilobes and quadrilobes. 20% deviation envelope.

701



702

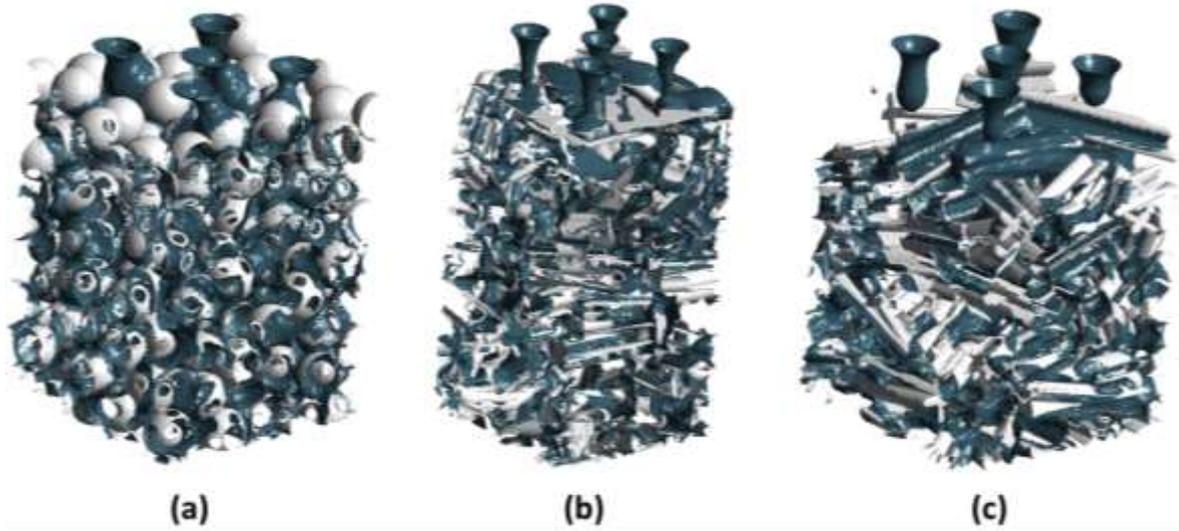
703

Figure 6: Parity diagram of two-phase pressure drop comparison between (Boyer et al. 2007) and CFD for  $V_{S,L} = 0.2 \text{ cm/s}$  (small markers),  $V_{S,L} = 0.5 \text{ cm/s}$  (medium markers),  $V_{S,L} = 0.8 \text{ cm/s}$  (large markers) and  $V_{S,G} = 5, 10, 20 \text{ cm/s}$ . Sphericity factors are respectively  $\Psi = 1$ ,  $\Psi = 0.81$  and  $\Psi = 0.42$  for spheres, trilobes and quadrilobes. 20% deviation envelope.

704

705

706

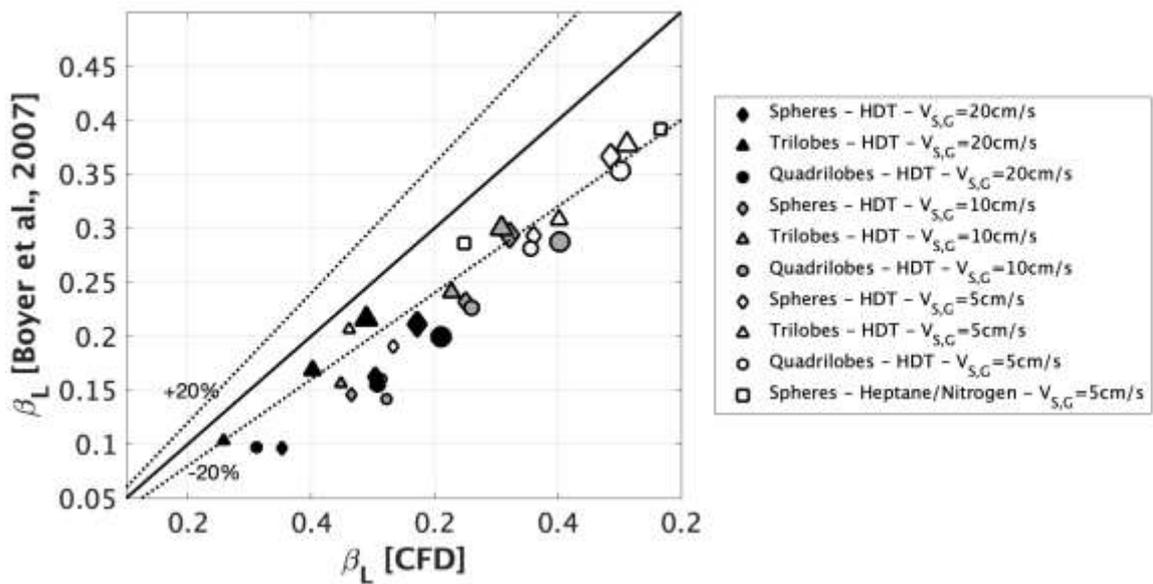


707

708

709

Figure 7: HDT flow through the different numerical domains: (a) Spheres, (b) Trilobes and (c) Quadrilobes at  $V_{S,L} = 0.5 \text{ cm/s}$  and  $V_{S,G} = 10 \text{ cm/s}$ . The liquid is represented by the blue color.



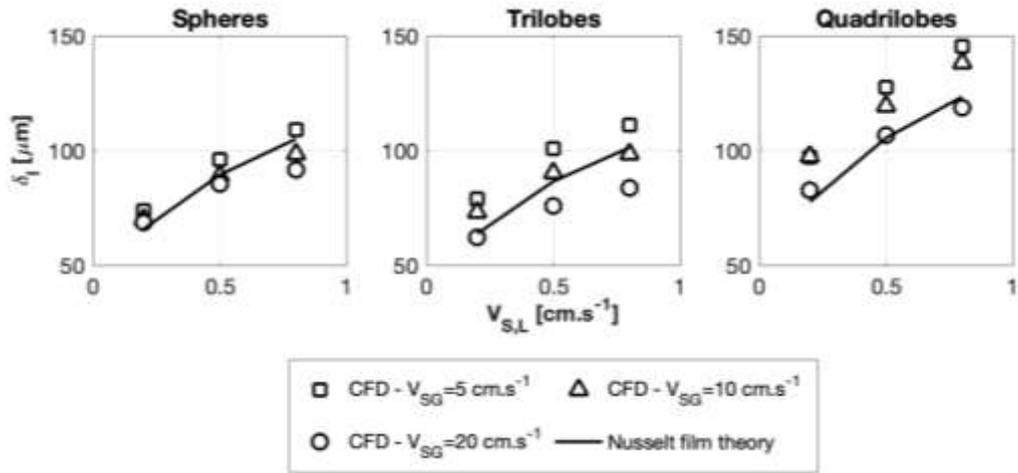
710

711

712

713

Figure 8: Parity diagram of liquid saturation comparison between (Boyer et al. 2007) and CFD for  $V_{S,L} = 0.2 \text{ cm/s}$  (small markers),  $V_{S,L} = 0.5 \text{ cm/s}$  (medium markers),  $V_{S,L} = 0.8 \text{ cm/s}$  (large markers) and  $V_{S,G} = 5, 10, 20 \text{ cm/s}$ . 20% deviation envelope.



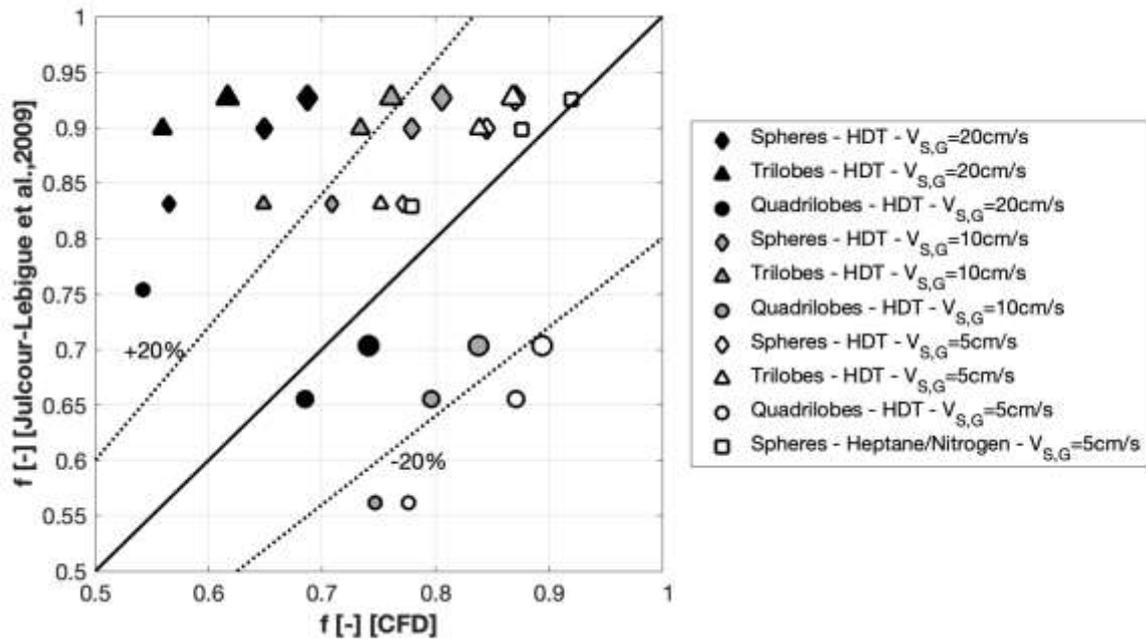
714

715

Figure 9: Comparison of Nusselt [1916] liquid film thickness to CFD results for spheres, trilobes and quadrilobes at

716

$V_{S,L} = 0.2, 0.5, 0.8 \text{ cm/s}$  and  $V_{S,G} = 5, 10, 20 \text{ cm/s}$ .



717

718

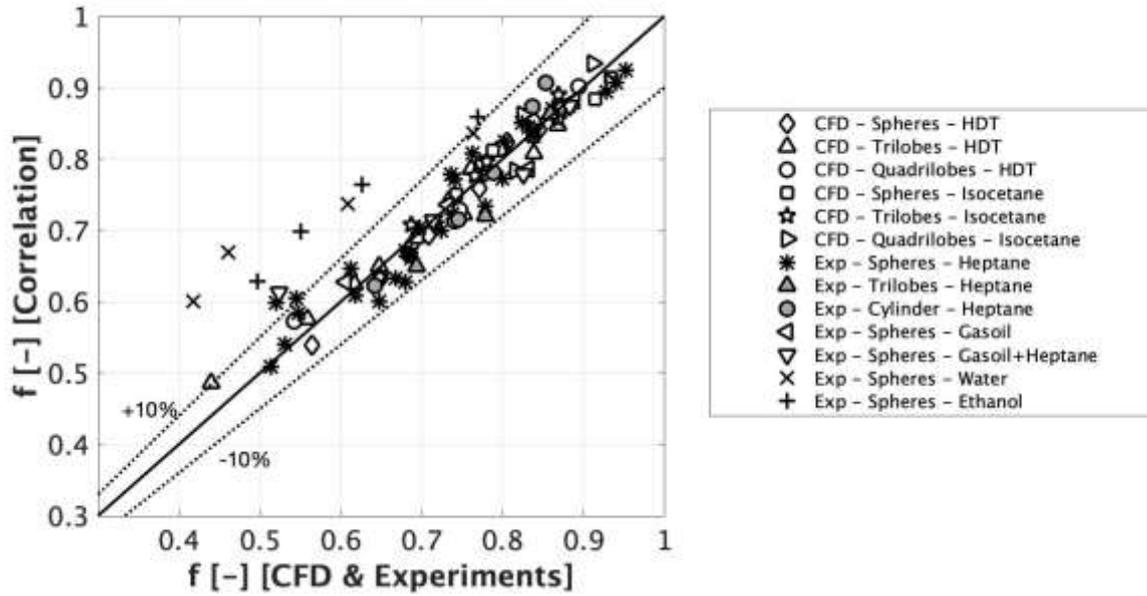
Figure 10: Wetting efficiency parity diagram. Comparison between (Julcour-Lebigue et al., 2009) and CFD for  $V_{S,L} =$

719

$0.2 \text{ cm/s}$  (small markers),  $V_{S,L} = 0.5 \text{ cm/s}$  (medium markers),  $V_{S,L} = 0.8 \text{ cm/s}$  (large markers) and  $V_{S,G} =$

720

$5, 10, 20 \text{ cm/s}$ . 20% deviation envelope.



721

722

Figure 11: Wetting efficiency parity diagram. Comparison between the correlation derived in this work (Equation (15)), CFD results and former experimental results of Julcour-Lebigue et al. (2009a). Dotted lines represent the  $\pm 10\%$

723

724

error envelope.

725

726

727

728

729

730

731

732

733

734 **Tables**735 **Table 1: Characteristics of particles and packings.**

Particle shape	Particle's equivalent diameter $d_e$ [mm]	Polydispersity		Bed porosity
		Mean length [mm]	Standard deviation [mm]	
Sphere	2.00	---		0.372
Trilobe	1.80	4.77	0.377	0.377
Quadrilobe	1.95	4.77	0.628	0.628

736

737 **Table 2 – Physical properties of the fluids composing the two-phase systems.**

System	Property	Liquid	Gas
Heptane-Nitrogen System	Density ( $\text{kg/m}^3$ )	683.8	1.249
	Viscosity (cP)	0.39	0.017
	Surface Tension ( $\text{mN/m}$ )		20
Hydrotreatment System (Hydrocarbon-hydrogen)	Density ( $\text{kg/m}^3$ )	750	15
	Viscosity (cP)	0.11	0.013
	Surface Tension ( $\text{mN/m}$ )		3.44
Isohexadecane-Nitrogen System	Density ( $\text{kg/m}^3$ )	784.4	1.249
	Viscosity (cP)	3.75	0.017
	Surface Tension ( $\text{mN/m}$ )		24.2

738

739 **Table 3 - Mesh sizes and wetting efficiencies for  $V_{SL} = 0.5 \text{ cm/s}$  and  $V_{SG} = 5 \text{ cm/s}$ .**

Mesh Resolution	Coarse Mesh	Medium Mesh	Fine Mesh	Very Fine Mesh
Number of cells	1.7M	3.4M	4.3M	11.2M
Wetting Efficiency	0.66	0.71	0.73	0.74

740

741

Table 4 - Particle shapes, bed porosity, liquid and gas superficial velocities tested to validate the model.

Particle shape	Bed void fraction $\varepsilon_B$	$V_{S,L}$ [cm/s]	$V_{S,G}$ [cm/s]
Sphere	0.372	0.2	5, 10, 20
		0.5	5, 10, 20
		0.8	5, 10, 20
Trilobe	0.377	0.2	5, 10, 20
		0.5	5, 10, 20
		0.8	5, 10, 20
Quadrilobe	0.628	0.2	5, 10, 20
		0.5	5, 10, 20
		0.8	5, 10, 20

742

743

Table 5 - Standard deviations obtained with the different dimensionless numbers group combinations.

Case Number	$C_1$	Exponents $C_i$ for $i>1$			Standard deviation $\sigma$ [%]
		$Re_G$	$Fr_G$	$Ga_G$	
1	-0.092	0.283	0	0	7.40%
2	-0.437	0	0.367	0	4.98%
3	0.074	0	0	0.141	8.72%
4	-0.459	-0.006	0.362	0	4.98%
5	-0.451	0	0.359	-0.003	4.98%
6	-0.451	0.718	0	-0.362	4.98%
7	-0.451	0.118	0.300	0.062	4.98%

744

745

Table 6 - Shape factor values for each particle shape.

Shape	Spheres	Trilobes	Quadrilobes
$\phi$	1.00	0.91	2.41

746

747 Table 7 - Standard deviation obtained by the model of Julcour-Lebigue et al. (2009a) and the new developed  
 748 correlation for three shape factor expressions.

	Julcour-Lebigue et al. (2009a) correlation	Shape factor for the new correlation		
		Eq. (13)	Eq. (15)	Eq. (17)
<b>Standard deviation</b>	13.18%	4.99%	6.04%	6.04%

749

750 Table 8 - Range of physical properties of gases and liquids, geometric properties of packings and operating conditions.

<b>Physical properties of gases and liquids used</b>	
Liquid density, $\rho_L$	$680 \text{ kg/m}^3 \leq \rho_L \leq 830 \text{ kg/m}^3$
Gas density, $\rho_G$	$1.249 \text{ kg/m}^3 \leq \rho_G \leq 15 \text{ kg/m}^3$
Liquid viscosity, $\mu_L$	$0.11 \times 10^{-3} \text{ Pa.s} \leq \mu_L \leq 3.75 \times 10^{-3} \text{ Pa.s}$
Gas viscosity, $\mu_G$	$1.3 \times 10^{-5} \text{ Pa.s} \leq \mu_G \leq 1.7 \times 10^{-5} \text{ Pa.s}$
Surface tension, $\sigma_L$	$3.44 \times 10^{-3} \text{ N/m} \leq \sigma_L \leq 28.1 \times 10^{-3} \text{ N/m}$
<b>Geometric properties of packings used</b>	
Equivalent diameter, $d_p$	$1.44 \text{ mm} \leq d_p \leq 7 \text{ mm}$
Bed porosity, $\varepsilon_B$	$0.367 \leq \varepsilon_B \leq 0.611$
Particle shape	Spheres, trilobes and quadrilobes
Packing specific surface area, $a_p$	$516 \text{ m}^{-1} \leq a_p \leq 2604 \text{ m}^{-1}$
<b>Operating conditions</b>	
Liquid mass flow flux, $L$	$1.34 \text{ kg/m}^2 \text{ s} \leq L \leq 60.1 \text{ kg/m}^2 \text{ s}$
Gas mass flow flux, $G$	$0 \text{ kg/m}^2 \text{ s} \leq G \leq 3 \text{ kg/m}^2 \text{ s}$
Liquid superficial velocity, $V_{S,L}$	$0.2 \text{ cm/s} \leq V_{S,L} \leq 0.8 \text{ cm/s}$
Gas superficial velocity, $V_{S,G}$	$5 \text{ cm/s} \leq V_{S,G} \leq 20 \text{ cm/s}$

751