Computational Investigation of Liquid Holdup and Wetting Efficiency Inside Trickle Bed Reactors with Different Catalyst Particle Shapes

Hao Deng 1,2,3,†, Baoqi Guo 1,2,3,†, He Dong 1,2,3, Cheng Liu 1,2,3 and Zhongfeng Geng 1,2,3,∗

1 Key Laboratory for Green Chemical Technology of Ministry of Education, Tianjin University, Tianjin 300072, China; denghao@tju.edu.cn (H.D.); guobaoqi4521@163.com (B.G.); donghe@tju.edu.cn (H.D.); liuc@tju.edu.cn (C.L.)
2 R&D Center for Petrochemical Technology, Tianjin University, Tianjin 300072, China
3 Collaborative Innovation Center of Chemical Science and Engineering, Tianjin 300072, China
∗ Correspondence: zfgeng@tju.edu.cn; Tel.: +86-22-27406119
† These authors contributed equally to this article.

Abstract: Liquid holdup and wetting efficiency are essential parameters for design of trickle bed reactors. Both parameters play an important role in reactor performance including pressure drop, conversion, and heat transfer. Empirical formulas are usually employed to calculate liquid holdup and wetting efficiency. However, factors such as particle shape and the wetting ability of liquid on the particle surface are not described clearly in traditional formulas. In this paper, actual random packing was built by DEM and CFD simulations were performed to investigate the factors affecting liquid holdup and wetting efficiency in trickle bed reactors, including particle shape, surface tension, contact angle, liquid viscosity, liquid density, liquid, and gas superficial velocity. Detailed fluid flow and liquid-solid interaction were described by VOF model. Four different particle shapes were investigated. It showed the particle shape has great effect and the 4-hole cylinder packing gained both highest liquid holdup and wetting efficiency. The overall simulations gave a detailed description of phase interactions and fluid flow in the voids between catalyst particles and these results could give further guidance for the design and operation of trickle bed reactors.

Keywords: liquid holdup; wetting efficiency; trickle bed reactor; CFD; VOF

1. Introduction

The gas-liquid-solid three-phase catalytic reaction is a common reaction type in chemical engineering. Many kinds of gas-liquid-solid three-phase reactors are employed in the industry, among which trickle bed reactors (TBRs) are widely used in fields such as petroleum, petrochemical, fine chemicals, and biochemical industries. They offer many advantages such as easy operation, high solid loading, and operation pressure [1]. In a catalyst packed bed, gas and liquid reactants can flow either concurrently downward or in opposite directions. The liquid phase on particles surface appears as a film or droplet. The phase interaction between gas, liquid, and solid in TBR makes the hydrodynamic phenomena quite complicated, which is usually investigated by experimental method [2–4]. When gas and liquid flow concurrently downward through a packed bed, four flow regimes were found. They are trickle flow, pulse flow, spray flow, and bubbly flow [5]. The characteristics of these flow regimes are different, and they affect the hydrodynamic parameters of TBRs significantly. Among these parameters,
liquid holdup and wetting efficiency are of great importance, which give descriptions of the liquid distribution. Liquid holdup $\varepsilon_L$ measures the quantity of liquid inside TBRs:

$$\varepsilon_L = \frac{\text{volume of liquid}}{\text{volume of TBR}}$$  \hspace{1cm} (1)

Another description of liquid quantity inside TBRs is the liquid saturation:

$$\beta_L = \frac{\text{volume of liquid}}{\text{volume of voids between particles inside TBR}}$$  \hspace{1cm} (2)

And the wetting efficiency is the percentage of wetting area of catalyst surface:

$$\eta_L = \frac{\text{wetting area of particles}}{\text{particles surface area}}$$  \hspace{1cm} (3)

The overall liquid holdup and wetting efficiency are usually calculated by empirical formulas. However, local liquid distribution phenomenon on the particle scale such as flow maldistribution, channeling and catalyst wetting can also significantly affect the overall reactor performance (e.g., pressure drop, catalyst use, and reactant conversion). The rigorous mathematical models for calculating them are still tough [6]. The liquid flow textures in a bed consist of several features: liquid film flow, rivulets over the particles, pendular structures, liquid-filled channels, and liquid-filled pockets [7]. They have various particle surface liquid coverages and perform differently with different gas-liquid catalytic reactions (liquid-limited or gas-limited). Also, the maldistribution of liquid caused by variation of voidage can lead to the local hot spot formation, causing temperature runaway and operation safety problems. The flow phenomenon mentioned above is significantly affected by factors such as particle shape, wetting ability of liquid on the particle surface, which are not described clearly in traditional formulas. Since the local liquid distribution has a significant impact on reactor performance, better models are needed for its accurate prediction. They will significantly aid the predictions of local temperatures and local concentration distributions, facilitating better reactor design [8]. Therefore, it is very important to accurately estimate the local liquid distribution on the particle scale in TBRs for further design and operation.

In recent years, computational fluid dynamics (CFD) has become a powerful method for investigating fluid behaviors inside TBRs and has attracted a lot of attention [5,9–11]. To simulate multiphase flow inside TBRs with CFD, many multiphase models have been employed. The Eulerian-Eulerian model for gas-liquid-solid flows is the most commonly used CFD model to predict the dynamic behavior of TBRs [12]. However, the Eulerian-Eulerian approach does not accomplish interface tracking by solving a continuity equation of the volume fraction of one (or more) phases, and for this reason it is unable to capture the wetting characteristics of the gas-liquid interface in the TBRs [13]. This drawback is more significant when we perform simulations with random packings, since a random packing has lots of tiny voids where surface tension has a great effect. Volume of fluid (VOF) model is one of the best implicit free-surface reconstruction methods. The VOF model is a surface-tracking technique applied to a fixed Eulerian mesh when the locus of the interface between two or more immiscible fluids is of interest. The main hurdles of using VOF rest on preserving a sharp boundary between the immiscible fluids and the computation of surface tension [14]. The velocity field and bubble profile in a vertical gas-liquid slug flow inside the capillaries has been modeled with a VOF technique, and the result was found to be in good agreement with published experimental measurements [15]. In the past two decades, the VOF model has been predominately practiced in the field of CFD simulation of TBRs [6,13,16–20].

Many researchers are devoted to conducting simulation research about TBRs. Jindal et al. performed Eulerian multi-fluid simulations to study the effect of bed characteristics of local liquid spreading in a TBR [10]. Lu et al. developed a new porous media model for CFD simulations of the
gas-liquid two-phase flow in rotating packed beds [11]. However, these papers either modeled solid particles as a continuum phase or regarded the whole simulation region as a porous media. They only calculated the fluid-solid interactions through specific methods, without considering the influence of the actual particle shape on the fluid flow. Several authors made efforts to perform particle-resolved TBR simulations. Lopes et al. proposed a high-resolution CFD approach using discrete particles and designed a TBR with regular positioned catalyst particles for the Euler-Euler [21] and Eulerian VOF simulations [13], and studied the catalytic wet oxidation of phenolic wastewaters in their later studies [22,23]. Du et al. performed CFD simulations on regular particles arrangement within a rectangular domain to investigate the local wetting behavior in a TBR [6,16]. Simulations performed by both groups modeled particles with regular configuration and fixed distance between each other, which is difficult to achieve in a real reactor where catalyst particles are packed randomly, and the particle’s shape can be complex such as a cylinder or hollow cylinder. Besides, their grid resolution are not high enough to give clear descriptions of the liquid flow configuration on the discrete particle surfaces.

In this paper, detailed packing geometries with different particle shapes by discrete element method (DEM). In addition, the effects of factors such as surface tension and contact angle are discussed, which are hardly shown in empirical formulas. The CFD simulation was employed with the volume of fluid (VOF) method to investigate the factors affecting liquid holdup and wetting efficiency in a trickle bed reactor. This research can give guidance in trickle bed reactor design and operation in the future.

2. Models and Methods

2.1. Governing Equations

2.1.1. DEM Model

To construct the actual geometry of a random packed bed, discrete element method (DEM) was employed to generate random packing recently. The DEM and CFD methods were developed and show better performance in many applications, especially in fixed bed reactor simulations. Eppinger et al. introduced the DEM-CFD meshing method for fixed beds consisting of monodisperse spherical particles [24]. Wehinger et al. combined particle packing structure with detailed reaction mechanisms of catalytic dry reforming of methane over rhodium and studied the interaction between chemical kinetics and transport of momentum, heat, and mass [25]. Zhang et al. studied the flow and heat transfer in fixed beds considering the packing structures of cylindrical particles and low tube to particle diameter ratios [26].

In this research, the DEM method was used to generate actual geometry of random packings with different particle shapes. The distinct characteristic of DEM is that inter-particle contact forces are included in the equations of motion. The size, shape, and orientation of each particle are taken into account. The governing equation describing particle motion is:

\[ m_p \frac{dv_p}{dt} = F_s + F_b \]  \hspace{1cm} (4)

where \( m_p \) mass of particle, \( v_p \) is particle velocity, \( t \) is time, \( F_s \) and \( F_b \) is surface and volume force respectively. \( F_b \) could be calculated by:

\[ F_b = F_g + F_c \]  \hspace{1cm} (5)

where \( F_g \) is gravity, \( F_c \) is the interaction force between particles and between particles and wall:

\[ F_g = m_p g \]  \hspace{1cm} (6)

\[ F_c = \sum_{contacts} F_{cm} \]  \hspace{1cm} (7)
$F_{cm}$ is contact force which could be calculated as:

$$F_{cm} = F_n + F_t$$

(8)

where $F_n$ is the normal and $F_t$ is the tangential force component:

$$F_n = -K_n d_n - N_n v_n$$

(9)

If $|K_t d_t| < |K_n d_n| C_{fs}$, $F_t$ is defined as:

$$F_t = -K_t d_t - N_t v_t$$

(10)

Otherwise:

$$F_t = \frac{|K_n d_n| C_{fs} d_t}{|d_t|}$$

(11)

where $K_n$ and $K_t$ are the normal and tangential spring constant, $N_n$ and $N_t$ are the normal and tangential damping, $d_n$ and $d_t$ are overlaps in the normal and tangential directions at the contact point respectively, $C_{fs}$ is the static friction coefficient. $N_n$ and $N_t$ could be calculated as follows:

$$N_n = 2 N_{ndamp} \sqrt{K_n M_{eq}}$$

(12)

$$N_t = 2 N_{tdamp} \sqrt{K_t M_{eq}}$$

(13)

$N_{ndamp}$ and $N_{tdamp}$ are normal and tangential damping coefficient respectively, $M_{eq}$ is the equivalent particle mass. These two damping coefficient could be defined as follows:

$$N_{ndamp} = \frac{- \ln(C_{n \text{rest}})}{\sqrt{\pi^2 + \ln(C_{n \text{rest}})^2}}$$

(14)

$$N_{tdamp} = \frac{- \ln(C_{t \text{rest}})}{\sqrt{\pi^2 + \ln(C_{t \text{rest}})^2}}$$

(15)

where $C_{n \text{rest}}$ and $C_{t \text{rest}}$ are the normal and tangential coefficients of restitution respectively. The rolling of particles (especially non-sphere particles) has great impact on the motion of particles. The rotational momentum equation is given as:

$$I_p \frac{d\omega_p}{dt} = M_b + M_c$$

(16)

where $I_p$ is the particle moment of inertia, $\omega_p$ is particle angular velocity, $M_b$ is the moment that acts on the particle (rotational drag), $M_c$ is the moment that acts on an individual particle due to contact force, which in turn acts on the particle at a point other than the particle center of gravity:

$$M_c = \sum_{\text{contacts}} (r_c \times F_{cm} + M_{cm})$$

(17)

where $r_c$ is the vector from the particle center of gravity to the contact point and $M_{cm}$ is the moment that act on the particle from contact models (rolling resistance).

2.1.2. VOF Model

The VOF model assumes that all immiscible fluid phases present in a control volume share their velocity, pressure, and temperature fields. Therefore, the same set of basic governing equations describing momentum, mass, and energy transport in a single-phase flow is solved. It is necessary to
know the volume fraction $\alpha_i$ of phase $i$ (liquid or gas), and the conservation equation that describes the transport of $\alpha_i$ is:

$$\frac{\partial (\alpha_i \rho_i)}{\partial t} + \nabla \cdot (\alpha_i \rho_i \mathbf{v}) = 0$$

(18)

where $\rho_i$ is the density, $t$ is time and $\mathbf{v}$ is the velocity. The momentum equation is given as:

$$\frac{\partial (\rho \mathbf{v})}{\partial t} + \nabla \cdot (\rho \mathbf{v} \otimes \mathbf{v}) = -\nabla p + \nabla \cdot \left[ \mu \left( \nabla \mathbf{v} + \nabla \mathbf{v}^T \right) \right] + \rho \mathbf{g} + \mathbf{F}$$

(19)

where $p$ is pressure, $\mathbf{g}$ is gravity and $\mathbf{F}$ is the resultant of other body forces. The physical properties are calculated as functions of the physical properties of its constituent phases and their volume fractions:

$$\rho = \sum_i \rho_i \alpha_i$$

(20)

$$\mu = \sum_i \mu_i \alpha_i$$

(21)

The surface tension force is a tensile force tangential to the interface separating the two fluids. It works to keep the fluid molecules at the free surface in contact with the rest of the fluid and shows remarkable effects on liquid behaviors in TBRs. The surface tension force $f_\sigma$, calculation method named continuum surface force (CSF) model was proposed by Brackbill et al. as the following [27]:

$$f_\sigma = \sigma \kappa \mathbf{n}$$

(22)

$\sigma$ is the surface tension coefficient and the vector normal to the interface $\mathbf{n}$ is calculated using the smooth field of the phase volume fraction $\alpha_i$:

$$\mathbf{n} = \nabla \alpha_i$$

(23)

The curvature of the interface $\kappa$ can therefore be expressed in terms of the divergence of the unit normal vector as follows:

$$\kappa = -\nabla \cdot \frac{\nabla \alpha_i}{|\nabla \alpha_i|}$$

(24)

Wall adhesion was considered by adjusting normal vector with contact angle:

$$\mathbf{n} = \mathbf{n}_{\text{wall}} \cos \theta + \mathbf{n}_1 \sin \theta$$

(25)

where $\theta$ is the contact angle, $\mathbf{n}_{\text{wall}}$ is the unit normal vector of solid wall, and $\mathbf{n}_1$ is the unit vector inside the wall normal to the solid-liquid-gas contact line.

2.2. Numerical Setup

Two parts of simulations were performed in this research. To verify the accuracy of the simulation framework, a simulation of a droplet falling on a sphere particle was carried out according to Du et al.’s research [26], and the outcome was compared with their results of both experimental and numerical simulation. Then, a packed bed structure was generated by DEM method and factors affecting the liquid distribution were investigated in the complex region. The properties of gas and liquid employed in verify simulations and CFD simulations referred to air and water under standard conditions and are given in Table 1.

All simulations mentioned in this research were conducted with polyhedral mesh. The reason to choose it is that polyhedral cells are relatively easy and efficient to build, requiring no more surface preparation than the equivalent tetrahedral mesh. Besides, they also contain approximately five times fewer cells than a tetrahedral mesh for a given starting surface. It provides great advantages to the
simulation regions, since it contains extremely complex geometry and requires large number of cells to describe the shape of the random packing. It will help us to build cells with high quality and speed up the burdensome simulations. All these simulations were implemented with the commercial software STAR-CCM+ 12.02 [28].

Table 1. Gas and liquid properties employed in both verify and DEM-CFD simulations.

| Gas and Liquid Properties      | Value                          |
|-------------------------------|-------------------------------|
| Liquid viscosity $\mu_L$ (Pa·s) | $8.8871 \times 10^{-4}$       |
| Gas viscosity $\mu_G$ (Pa·s)   | $1.85508 \times 10^{-5}$      |
| Liquid density $\rho_L$ (kg/m³) | 1000                          |
| Gas density $\rho_G$ (kg/m³)   | 1.225                         |
| Surface tension coefficient $\sigma$ (N/m) | 0.072                      |
| Contact angle $\theta$        | 45°                           |

2.2.1. Validation Model

Du conducted the experiment and performed the 2D VOF simulation which received satisfactory flow patterns comparable with the experiment results [19]. However, the 2D simulation was closer to a situation where a cylindrical drop of infinite length falls on an infinite-high cylinder, which is far from reality. Therefore, to better verify the accuracy of this method, the 3D simulations were performed.

The mesh plot of 3D verification model and measuring positions are shown in Figure 1. The simulation scales are the same as those in Du’s experiments [19]. The validation experiment was conducted under atmospheric pressure and at a temperature of 273 K. A particle of 10 mm in diameter was positioned in a cylinder region whose diameter and length are 20 mm and 25 mm respectively. A water droplet of 4 mm in diameter was patched 5 mm above the particle and slightly offset from the central axis (about 2.82 mm). For the region within 17 mm of the particle and droplet surface, meshes with the highest resolution were used (0.07 mm for 3D cases). For the other region, the largest-scale nonstructural meshes were adopted. The top entrance was defined as the velocity inlet for the gas phase and the exit was defined as pressure outlet conditions. The time step used in the simulation was $1 \times 10^{-5}$ s, and the initial velocity for the liquid droplet was set to 0 so that the droplet fell down only by gravity. The simulated results were recorded every 5 ms, and the liquid film thickness was measured at different positions of the particle surface at different time intervals.

Figure 1. Mesh plot of 3D verification model and different measuring positions.
2.2.2. CFD Simulation

The mesh of the simulated TBR model is shown in Figure 2. The 6 mm × 6 mm × 9 mm TBR model was defined as the simulation region. Four different particle shapes were considered in this research, including sphere, cylinder, single-hole cylinder, and 4-hole cylinder. Table 2 shows the geometric size data of four kinds of particles.

![Figure 2. Illustration of four simulated packing meshes including sphere (a), cylinder (b), single-hole cylinder (c), and 4-hole cylinder (d).](image)

| Table 2. Geometric size of four kinds of particles. |
| Shape | Characteristic Size (mm) | Shape | Characteristic Size (mm) |
|-------|--------------------------|-------|--------------------------|
| Sphere | Diameter 2 | Cylinder | Height 2.27 |
| | Height 2.27 | | Diameter 1.36 |
| 4-hole cylinder | Diameter 1.36 | Single-hole cylinder | Height 2.27 |
| | Inner holes diameter 0.34 | | Diameter 1.36 |
| | Inner holes center distance 0.68 | | Inner hole diameter 0.68 |

The diameter of the sphere was 2 mm. The surface area of the cylinder particle was the same as the sphere, and the ratio of diameter to height was 3/5. The diameters of the inner holes of the single-hole cylinder and 4-hole cylinder are half and a quarter of the cylinder diameter, respectively.

About 56 random spheres and 77 cylindrical particles inside rectangular domains were generated through DEM method. The liquid was introduced into the TBR from the top through the four tubes with diameter $D_{in} = 0.5$ mm and length $L_{in} = 1$ mm, which were located on the top forming a 3 mm × 3 mm square. Different numbers of inlets were tested, including 1, 4, and 9 inlets on the top. We found that there was no significant difference in simulation results between different inlet numbers. Therefore, the middle number of 4 inlets was selected. The residual area of the top was the gas inlet. The gas and liquid inlets were defined as velocity inlets and the region bottom was a pressure outlet. Four cross sections were set as symmetrical planes. The VOF model suffer from the generation of spurious currents near the interface. However, to get convergent simulation results, there is no treatment about spurious currents in this research. The time step was $2 \times 10^{-4}$ s and the mesh numbers were 269,516, 1,112,159, 1,395,718, and 1,604,862, respectively. Two high-performance compute cluster with 8 CPU-cores were used to perform the simulations. The calculation time of four different models are 9.0, 48.0, 57.3, and 65.9 h respectively.

The independence of time step and mesh number was confirmed before the decisions were made. Except for the investigation of particle shape's influence, the investigations of the factors were implemented on a random 4-hole cylinder packing. All simulations were conducted in the theoretical trickle flow regime. The volume of liquid and the wetted area of each case are monitored. At first both two quantities increased fast, while after some time they stopped significant increase and began to fluctuate. After that the result data was calculated by time-averaging. In the following chapters, only
the investigated factors were changed, and the rest of fluid properties were kept constant as shown in Table 1.

3. Results and Discussion

3.1. Method Validation

The comparison of snapshots is shown in Figure 3. It can be seen that 3D simulations gave more accurate results for they described the extreme thin liquid film on the top of the particle at 20 ms and 25 ms better. This is due to the fact that the 3D region allowed liquid to flow along the particle surface in various directions. These results proved that the simulation framework was capable of describing the behavior of liquid on solid particles. However, it could be found that some differences exist between the reported experiment and 3D simulation results.

Figure 3. Comparisons of snapshots between experiment [19] and VOF simulations of a droplet falling.

Table 3 shows the thickness of liquid film on particle surface of different positions. Dry surface and unmeasurable thin film are ignored in the table. It can be seen there are still several significant difference between reported experiment and 3D simulation. Most of thicknesses in the experiment are larger than those in 3D simulations, and the wetting of particle surface is faster in the experiment. All these differences could be concluded from two reasons. First the droplet in simulation is perfect sphere while in experiment is nearly ellipsoidal, which is larger in volume. Therefore the thickness of liquid film and flowing velocity are larger in experiment. Second, it can be seen that in the experiment there is a droplet hanging on the bottom of the particle at t = 0 ms, which means the particle was wetted. However, the particle surface in the simulation was totally dry. This could lead to the differences between the experiment and simulation.
Table 3. Thickness of liquid film on particle surface of different positions.

| t (ms) | 0° | 20° | 40° | 60° | 80° | 100° |
|--------|----|-----|-----|-----|-----|------|
| 10 Exp. | 1.30 | 3.79 | 1.82 | 1.33 |     |      |
| Sim.    | 0.23 | 3.35 | 2.87 |     |      |      |
| 15 Exp. | 1.25 | 0.85 | 1.22 | 1.56 | 2.05 |      |
| Sim.    | 0.49 | 2.94 | 2.14 | 0.95 |     |      |
| 20 Exp. | 0.09 |     | 0.90 | 1.58 | 1.89 |      |
| Sim.    | 0.62 | 0.47 | 0.78 | 0.85 | 1.11 |      |
| 25 Exp. |     | 1.21 | 2.44 | 2.56 |     |      |
| Sim.    | 0.81 |     | 0.52 | 1.22 |     |      |

3.2. Factors Investigation

3.2.1. Catalyst Particle Shape

In TBRs, the voids formed between particles affect the fluid flow structure significantly hence affect the liquid holdup and wetting efficiency. The scope is limited when altering the local characteristics of packing/voids, but the manipulation of particle size and shape may allow some degree of control [1]. Packings formed by particles of different shape have different porosities and specific surface areas, and the different flow channels of various complexity have a great influence on the distribution of the flow field. Here four particle shapes (sphere, cylinder, single-hole cylinder, and 4-hole cylinder) were investigated for their characteristics of liquid holdup and wetting efficiency.

Characteristic data of three kinds of random packings was given in Table 4, and the liquid distributions inside four packings are shown in Figure 4. It can be clearly seen that the liquid inside four packings tended to accumulate in the inner holes and voids between particles. The cylinder packing had the lowest porosity liquid holdup among all. The difference in liquid holdup between spheres and cylinders was not significant since the contact form between cylinder particles was more complex than spheres, including point contact, line contact, and surface contact. This caused a higher number and more complex shapes of voids between particles, from which the cylinder packing got a stronger ability of keeping liquid by capillary effect. However, cylinders got higher liquid saturation than spheres due to their lower porosity. Single-hole cylinders showed much higher liquid holdup and saturation than both spheres and cylinders. In addition, 4-hole cylinders had both the highest liquid holdup and saturation. It can be found that the inner holes in the 4-hole cylinder packing were easier to be filled with liquid than those in a single-hole cylinder packing, due to the smaller inner hole diameter. The larger number of holes made the 4-hole cylinder packing gain the highest porosity and improved the ability of trapping liquid significantly.

Table 4. Characteristic data of four kinds of random packing structures.

| Shape             | Porosity | Liquid Holdup | Liquid Saturation | Surface Area (m²) | Wetting Efficiency | Pressure Drop (Pa) |
|-------------------|----------|---------------|-------------------|-------------------|--------------------|-------------------|
| Sphere            | 0.55     | 0.1784        | 0.3246            | 4.49 × 10⁻⁴       | 0.5883             | 236.50            |
| Cylinder          | 0.51     | 0.1711        | 0.3329            | 6.20 × 10⁻⁴       | 0.6322             | 401.60            |
| Single-hole cylinder | 0.63 | 0.2272        | 0.3600            | 8.29 × 10⁻⁴       | 0.6289             | 344.19            |
| 4-hole cylinder   | 0.66     | 0.2633        | 0.3955            | 1.02 × 10⁻³       | 0.6566             | 351.00            |
wetting efficiency. The packing had the lowest surface area and wetting efficiency. For cylinders, single-hole cylinders, and 4-hole cylinders, the surface area increased with the rise of the inner hole number. However, their wetting efficiencies were comparable, as the small inner holes of hollow cylinders got wet easily due to the capillary effect. It can be concluded that sphere particles were suitable in situations which prefer low liquid holdup and wetting efficiency, while single-hole and 4-hole cylinders showed the opposite characteristics. Cylinder particles give lower liquid holdup, but higher wetting efficiency.

As for the pressure drop, the cylinder packing got the highest one due to the lowest porosity. Comparing cylinder and sphere packing, their liquid holdup and saturation were similar, but the lower porosity gave less free space for gas flow between cylinders, making a remarkable higher pressure drop. While for the single-hole and 4-hole cylinders, whose liquid holdups were much higher than sphere and cylinder packings, the high porosity provided a lot of space for gas flow even though most of them were filled with liquid. Therefore, their pressure drops were lower than the cylinder packing. However, the pressure drop in 4-hole cylinders packing was slightly higher than that in the single-hole cylinder.

Comparing these four kinds of particles, the 4-hole cylinder has both the highest liquid holdup and wetting efficiency, which are preferred by most of the gas-liquid-solid catalyzed systems. Therefore, the 4-hole cylinder was decided as the main investigation object in the following chapters.

3.2.2. Surface Tension Coefficient

Surface tension coefficient gives the measurement on the contractility of the liquid-gas interface. Surface tension is caused by cohesive forces between similar molecules and it shows distinct effect when liquid flows through tiny regions like voids between particles and inner holes of particles. The effect of surface tension coefficient on liquid holdup and wetting efficiency was investigated and the results are shown in Table 5 and Figure 5.

**Table 5.** Liquid holdup and wetting efficiency of three surface tension coefficient.

| Surface Tension Coefficient (N/m) | Liquid Holdup | Wetting Efficiency |
|----------------------------------|--------------|--------------------|
| 0.036                            | 0.2401       | 0.6008             |
| 0.072                            | 0.2633       | 0.6566             |
| 0.108                            | 0.2701       | 0.6687             |

Figure 4. Liquid distributions inside four packing structures ($v_G = 0.245 \text{ m/s}$, $v_L = 0.008 \text{ m/s}$).
It can be seen that some liquid in the 70° simulation tended to gather as a liquid drop (the circled position in Figure 6). This discovery of liquid holdup variation might be explained by the fact that liquid with a low contact angle tends to flow on particle surface as a thin film, making it harder to fill the voids and inner holes inside the packing. In this case, the gas gains more flow channels and it becomes easier for the liquid to leave the random packing, while liquid with a too high contact angle is less hydrophilic, which tends to gather into a liquid drop on surface due to its ability of silting channels and cohere with the particle surface.

3.2.3. Contact Angle

The contact angle describes the wetting ability of a liquid on a specific solid surface, which is affected by multiple factors including liquid component, solid component, and surface roughness. A higher contact angle means lower wetting ability, while a lower contact angle means that the liquid can spread easier on solid surfaces. The simulation results regarding the effect of contact angle are shown in Table 6 and Figure 6.

| Contact Angle (°) | Liquid Holdup | Wetting Efficiency |
|------------------|--------------|--------------------|
| 20               | 0.2503       | 0.6919             |
| 45               | 0.2633       | 0.6566             |
| 70               | 0.2488       | 0.5691             |

It can be seen that the wetting efficiency decreased significantly with the rise of contact angle because a higher contact angle resulted in accumulation rather than the spread of the liquid on particle surfaces. The same conclusion was made in other investigations on sphere particles. However, there was a noticeable phenomenon that 45° got the highest liquid holdup. Figure 6 shows that liquid in the 20° simulation tended to stay close to the particle surface. Most of the voids were free from liquids and some inner holes were not full of liquid. In the 45° simulation most of the space between particles was filled with liquid, while particles in the 70° were less surrounded by liquid for the weaker wetting ability. It can be seen that some liquid in the 70° simulation tended to gather as a liquid drop (the circled position in Figure 6). This discovery of liquid holdup variation might be explained by

Figure 5. Liquid distributions with three surface tension coefficients ($v_G = 0.245$ m/s, $v_L = 0.008$ m/s).
the fact that liquid with a low contact angle tends to flow on particle surface as a thin film, making it harder to fill the voids and inner holes inside the packing. In this case, the gas gains more flow channels and it becomes easier for the liquid to leave the random packing, while liquid with a too high contact angle is less hydrophilic, which tends to gather into a liquid drop on surface and slips down. So it can be concluded that a moderate contact angle can result in high liquid holdup due to its ability of silting channels and cohere with the particle surface.

Figure 6. Liquid distributions with different contact angles ($v_G = 0.245 \text{ m/s}, v_L = 0.008 \text{ m/s}$).

3.2.4. Liquid Viscosity

Liquid viscosity is another factor affecting liquid distribution. It affects fluid turbulence directly and thus affects the flow form of the liquid inside TBRs. Investigation results of the variation of liquid viscosity are given in Table 7, and the liquid distribution inside the packing is shown in Figure 7.

| Liquid Viscosity (Pa·s) | Liquid Holdup | Wetting Efficiency |
|-------------------------|---------------|--------------------|
| $4.4435 \times 10^{-4}$ | 0.2392        | 0.6237             |
| $8.8871 \times 10^{-4}$ | 0.2633        | 0.6566             |
| $13.3305 \times 10^{-4}$ | 0.2654        | 0.6518             |

Table 7. Liquid holdup and wetting efficiency of three liquid viscosities.

It can be clearly seen from Figure 7 that the quantity of liquid inside the packing increased with the rise of liquid viscosity. This is because the higher viscosity lead to a stronger liquid-solid shear, making it difficult for turbulence to develop, and resulted in a more uniform liquid flow on the particle surface. In high liquid viscosity cases the liquid-solid shear plays a greater role than the gas-liquid interactions [1], making the liquid harder to leave the particle surface and the whole packing. Film flow was easier to be found under high viscosity while rivulet flow can be observed under low viscosity [16], which is responsible for the increase in wetting efficiency. Differences between the first and the second was greater than that between the second and the third. For the wetting efficiency it increased by about 3% when the viscosity increased from $4.4435 \times 10^{-4}$ Pa·s to $8.8871 \times 10^{-4}$ Pa·s. However, a further increase of viscosity caused a slight drop in wetting efficiency, which did not vary distinctly. It shows the same tendency as liquid holdup, which means that the increase of liquid viscosity is more effective when the viscosity is low.
Thus, the liquid attained a higher speed and was able to leave the packing and the particle surface. The liquid density strengthened the gravity effect on the liquid flow, leading to a faster downward acceleration. The increase of liquid holdup and wetting efficiency decreased with the rise of liquid density. In Figure 8, the quantity of liquid in the voids between particles decreased noticeably, for more liquid trapped by surface tension and shear force was drawn out from the voids due to the increased gravity force. The increase of density strengthened the gravity effect on the liquid flow, leading to a faster downward acceleration. Thus, the liquid attained a higher speed and was able to leave the packing and the particle surface faster, resulting in a lower liquid holdup and wetting efficiency. Besides, it was reported that a higher liquid density makes rivulet flow easier than film flow [16], which was another reason for the lower wetting efficiency.

\[
\begin{align*}
\mu &= 4.435 \times 10^{-4} \text{ Pa·s} \\
\mu &= 8.871 \times 10^{-4} \text{ Pa·s} \\
\mu &= 13.3305 \times 10^{-4} \text{ Pa·s}
\end{align*}
\]

**Figure 7.** Liquid distributions with three liquid viscosities \((v_G = 0.245 \text{ m/s}, v_L = 0.008 \text{ m/s})\).

### 3.2.5. Liquid Density

The investigation result of influence on liquid holdup and wetting efficiency by liquid density is shown in Table 8, and the liquid distribution is shown in Figure 8. It can be found that the liquid holdup and wetting efficiency decreased with the rise of liquid density. In Figure 8, the quantity of liquid in the voids between particles decreased noticeably, for more liquid trapped by surface tension and shear force was drawn out from the voids due to the increased gravity force. The increase of density strengthened the gravity effect on the liquid flow, leading to a faster downward acceleration. Thus, the liquid attained a higher speed and was able to leave the packing and the particle surface faster, resulting in a lower liquid holdup and wetting efficiency. Besides, it was reported that a higher liquid density makes rivulet flow easier than film flow [16], which was another reason for the lower wetting efficiency.

\[
\begin{align*}
\rho_L &= 500 \text{ kg/m}^3 \\
\rho_L &= 1000 \text{ kg/m}^3 \\
\rho_L &= 1500 \text{ kg/m}^3
\end{align*}
\]

**Figure 8.** Liquid distributions with three liquid densities \((v_G = 0.245 \text{ m/s}, v_L = 0.008 \text{ m/s})\).
| Liquid Density (kg/m³) | Liquid Holdup | Wetting Efficiency |
|------------------------|--------------|--------------------|
| 500                    | 0.2694       | 0.6625             |
| 1000                   | 0.2633       | 0.6566             |
| 1500                   | 0.2543       | 0.6313             |

### 3.2.6. Liquid and Gas Velocity

The liquid and gas velocities inside TBRs have a great effect on the liquid distribution by affecting the inertia force. The simulation results of different liquid and gas superficial velocities are shown in Table 9, and the liquid distribution inside the spherical packing is shown in Figures 9 and 10. The increase of liquid velocity lead to a rise in both liquid holdup and wetting efficiency, while the increasing of gas velocity had the opposite effect.

| Liquid Superficial Velocity (m/s) | Gas Superficial Velocity (m/s) | Liquid Holdup | Wetting Efficiency |
|-----------------------------------|--------------------------------|--------------|--------------------|
| 0.002                             | 0.245                          | 0.2096       | 0.5686             |
| 0.005                             | 0.245                          | 0.2565       | 0.6339             |
| 0.008                             | 0.245                          | 0.2633       | 0.6566             |
| 0.008                             | 0.082                          | 0.3086       | 0.7163             |
| 0.008                             | 0.163                          | 0.2898       | 0.6881             |
| 0.008                             | 0.245                          | 0.2633       | 0.6566             |

In Figure 9 the accumulation of liquid inside the 4-hole cylinder packing is clearly shown. With increasing velocity, liquid tends to flow through more voids, then fills all possible space inside the TBR. It can be found that the quantity of liquid in $v_L = 0.002$ m/s was much less than $v_L = 0.005$ m/s and 0.008 m/s. Although the increase of liquid velocity gave rise to the liquid inertia force, it still cannot force liquid to leave the packing faster since the input quantity increased too. It is worth noticing that the increased liquid holdup from $v_L = 0.005$ m/s to 0.008 m/s was much less remarkable that that from 0.002 m/s to 0.005 m/s. It can be explained by the fact that most of the attainable free space was filled with liquid when $v_L = 0.005$ m/s and the increase of $v_L$ from 0.005 m/s to 0.008 m/s may have happened...
in the pulse flow regime, where the increase of liquid holdup with $v_L$ was less significant than which in the trickle flow regime.

The rise of the gas velocity brought a higher liquid-gas shear stress, making it easier for the liquid to leave the particle surface and the packing. The difference of liquid quantities shown in Figure 10 was remarkable, as the liquid quantity under $v_G = 0.245$ m/s was less than the other situations.

$$v_G = 0.082 \text{ m/s}$$
$$v_G = 0.163 \text{ m/s}$$
$$v_G = 0.245 \text{ m/s}$$

Figure 10. Liquid distributions with different gas superficial velocities ($v_L = 0.008$ m/s).

4. Conclusions

This paper employed CFD with a volume of fluid (VOF) method to implement high-precision simulations of the co-currently downward gas and liquid flow between catalyst particles inside TBRs. Traditional experimental methods have intrinsic limitations on liquid holdup and wetting efficiency measurement and most empirical equations which were concluded from these methods do not consider all effective factors such as the contact angle and surface tension coefficient. The detailed effective factors were investigated in this paper and remarkable findings are presented below.

(1) Four different catalyst particle shapes including sphere, cylinder, single-hole cylinder, and 4-hole cylinder were simulated to study their characteristics about liquid holdup and wetting efficiency. The 4-hole cylinder obtained both the highest liquid holdup and wetting efficiency. Spherical and cylindrical particles of the same volume have similar liquid holdups, but the wetting efficiency of cylindrical particles is higher than that of spherical particles. In addition, the liquid holdup of cylindrical particles increases with the number of inner holes (surface area), while the wetting efficiency does not change significantly.

(2) The variation of the contact angle and surface tension coefficient directly affects the accumulation and flow of liquid in voids between the particles inside the 4-hole cylinder packing, which further affect the liquid holdup and wetting efficiency. Both liquid holdup and wetting efficiency increase with the surface tension coefficient. The wetting efficiency decreases with the increase of contact angle, but a moderate contact angle can result in a high liquid holdup.

(3) Liquid viscosity and density affect the liquid holdup and wetting efficiency by affecting the liquid-solid shear force and gravity force, respectively. A higher viscosity or lower density of liquid can result in an increase of both liquid holdup and wetting efficiency.

(4) Gas superficial velocity affects the liquid holdup and wetting efficiency by affecting the inertial force and has the opposite effect on the surface tension and liquid-solid shear force. While liquid superficial velocity affects those two factors mainly by varying the liquid input quantity.
The increase in liquid velocity gives a rise in both liquid holdup and wetting efficiency, while the increase in gas velocity can reduce them.

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**Nomenclature**

- $C_{fs}$: static friction coefficient [-]
- $C_{n \text{ rest}}$: normal coefficients of restitution [-]
- $C_{t \text{ rest}}$: tangential coefficients of restitution [-]
- $D_{in}$: diameter of liquid inlet [m]
- $d_n$: overlaps in normal directions [-]
- $d_p$: diameter of particle [m]
- $d_t$: overlaps in tangential directions [-]
- $f_s$: surface tension force [N]
- $F$: resultant of other body forces [N]
- $F_b$: volume force [N]
- $F_c$: interaction force [N]
- $F_{cm}$: contact force [N]
- $F_g$: gravity [N]
- $F_n$: normal force component [N]
- $F_s$: surface force [N]
- $F_t$: tangential force component [N]
- $g$: gravitational acceleration [m/s$^2$]
- $G$: gas superficial mass velocity [kg/m$^2$·s]
- $L$: liquid superficial mass velocity [kg/m$^2$·s]
- $L_{in}$: length of liquid inlet [m]
- $I_p$: particle moment of inertia [kg·m/s]
- $K_n$: normal spring constant [-]
- $K_t$: tangential spring constant [-]
- $M_b$: rotational drag [kg·m/s]
- $M_c$: moment acts on individual particle [kg·m/s]
- $M_{cm}$: rolling resistance [kg·m/s]
- $M_{eq}$: equivalent particle mass [kg]
- $m_p$: mass of particle [kg]
- $n$: vector normal to the interface [-]
- $N_n$: normal damping [-]
- $N_{n \text{ damp}}$: normal damping coefficient [-]
- $N_t$: tangential damping [-]
- $N_{t \text{ damp}}$: tangential damping coefficient [-]
- $n_{\text{wall}}$: unit normal vector of solid wall [-]
- $n_t$: unit vector inside the wall normal to the solid-liquid-gas contact line [-]
- $p$: pressure [Pa]
- $t$: time [s]
- $v$: velocity [m/s]
- $v_p$: particle velocity [m/s]
- $v_L$: liquid superficial velocity [m/s]
- $v_G$: gas superficial velocity [m/s]
$r_c$ vector from particle center of gravity to contact point [-]
$\alpha_i$ volume fraction of phase $i$ [-]
$\beta_L$ liquid saturation [-]
$\epsilon$ porosity [-]
$\varepsilon_L$ liquid holdup [-]
$\eta_L$ wetting efficiency [-]
$\theta$ contact angle [$^\circ$]
$\kappa$ the curvature of the interface [-]
$\mu$ viscosity [Pa·s]
$\rho_i$ density [kg/m$^3$]
$\sigma$ surface tension coefficient [N/m]

References
1. Vivek, V.; Ranade, R.V.C.; Prashant, R.G. *Trickle Bed Reactors: Reactor Engineering & Applications*; Elsevier: Oxford, UK, 2011.
2. Sederman, A.J.; Gladden, L.F. Magnetic resonance imaging as a quantitative probe of gas-liquid distribution and wetting efficiency in trickle-bed reactors. *Chem. Eng. Sci.* 2001, 56, 2615–2628. [CrossRef]
3. Nemec, D.; Levec, J. Flow through packed bed reactors: 2. Two-phase concurrent downflow. *Chem. Eng. Sci.* 2005, 60, 6958–6970. [CrossRef]
4. Singh, B.K.; Jain, E.; Buwa, V.V. Feasibility of Electrical Resistance Tomography for measurements of liquid holdup distribution in a trickle bed reactor. *Chem. Eng. J.* 2019, 358, 564–579. [CrossRef]
5. Gunjal, P.R.; Kashid, M.N.; Ranade, V.V.; Chaudhari, R.V. Hydrodynamics of trickle-bed reactors: Experiments and CFD modeling. *Ind. Eng. Chem. Res.* 2005, 44, 6278–6294. [CrossRef]
6. Du, W.; Zhang, L.F.; Lv, S.; Lu, P.; Xu, L.; Wei, W. Numerical study of liquid coverage in a gas-liquid-solid packed bed. *Particuology* 2015, 23, 90–99. [CrossRef]
7. Maiti, R.; Khanna, R.; Nigam, K.D.P. Hysteresis in trickle-bed reactors: A review. *Ind. Eng. Chem. Res.* 2006, 45, 5185–5198. [CrossRef]
8. Dhanraj, D.I.A.; Buwa, V.V. Effect of capillary pressure force on local liquid distribution in a trickle bed. *Chem. Eng. Sci.* 2018, 191, 115–133. [CrossRef]
9. Kuzeljevic, Z.V.; Dudukovic, M.P. Computational Modeling of Trickle Bed Reactors. *Ind. Eng. Chem. Res.* 2012, 51, 1663–1671. [CrossRef]
10. Jindal, A.; Buwa, V.V. Effect of bed characteristics on local liquid spreading in a trickle bed. *AIChE J.* 2017, 63, 347–357. [CrossRef]
11. Lu, X.; Xie, P.; Ingham, D.B.; Pourkashanian, M. A porous media model for CFD simulations of gas-liquid two-phase flow in rotating packed beds. *Chem. Eng. Sci.* 2018, 189, 123–134. [CrossRef]
12. Iluta, I.; Larachi, F. Modelling the hydrodynamics of gas-liquid packed beds via slit models: A review. *Int. J. Chem. React. Eng.* 2005, 3, 27. [CrossRef]
13. Lopes, R.J.G.; Quinta-Ferreira, R.M. Numerical Simulation of Trickle-Bed Reactor Hydrodynamics with RANS-Based Models Using a Volume of Fluid Technique. *Ind. Eng. Chem. Res.* 2009, 48, 1740–1748. [CrossRef]
14. Wang, Y.N.; Chen, J.W.; Larachi, F. Modelling and simulation of trickle-bed reactors using computational fluid dynamics: A state-of-the-art review. *Can. J. Chem. Eng.* 2013, 91, 136–180. [CrossRef]
15. Taha, T.; Cui, Z.F. Hydrodynamics of slug flow inside capillaries. *Chem. Eng. Sci.* 2004, 59, 1181–1190. [CrossRef]
16. Du, W.; Zhang, J.X.; Lu, P.P.; Xu, X.; Wei, W.; Li, W.; Zhang, F. Advanced understanding of local wetting behaviour in gas-liquid-solid packed beds using CFD with a volume of fluid (VOF) method. *Chem. Eng. Sci.* 2017, 170, 378–392. [CrossRef]
17. Gunjal, P.R.; Ranade, V.V.; Chaudhari, R.V. Dynamics of drop impact on solid surface: Experiments and VOF simulations. *AIChE J.* 2005, 51, 59–78. [CrossRef]
18. Gunjal, P.R.; Ranade, V.V.; Chaudhari, R.V. Experimental and computational study of liquid drop over flat and spherical surfaces. *Catal. Today* 2003, 79, 267–273. [CrossRef]
19. Du, W.; Feng, D.S.; Xu, J.; Wei, W.S. Computational Fluid Dynamics Modeling of Gas-Liquid Two-Phase Flow around a Spherical Particle. *Chem. Eng. Technol.* **2013**, *36*, 840–850. [CrossRef]

20. Augier, F.; Koudil, A.; Royon-Lebeaud, A.; Muszynski, L.; Yanouri, Q. Numerical approach to predict wetting and catalyst efficiencies inside trickle bed reactors. *Chem. Eng. Sci.* **2010**, *65*, 255–260. [CrossRef]

21. Lopes, R.J.G.; Quinta-Ferreira, R.M. CFD modelling of multiphase flow distribution in trickle beds. *Chem. Eng. J.* **2009**, *147*, 342–355. [CrossRef]

22. Lopes, R.J.G.; Quinta-Ferreira, R.M. Assessment of CFD Euler-Euler method for trickle-bed reactor modelling in the catalytic wet oxidation of phenolic wastewaters. *Chem. Eng. J.* **2010**, *160*, 293–301. [CrossRef]

23. Lopes, R.J.G.; Quinta-Ferreira, R.M. Assessment of CFD-VOF Method for Trickle-Bed Reactor Modeling in the Catalytic Wet Oxidation of Phenolic Wastewaters. *Ind. Eng. Chem. Res.* **2010**, *49*, 2638–2648. [CrossRef]

24. Eppinger, T.; Seidler, K.; Kraume, M. DEM-CFD simulations of fixed bed reactors with small tube to particle diameter ratios. *Chem. Eng. J.* **2011**, *166*, 324–331. [CrossRef]

25. Wehinger, G.D.; Eppinger, T.; Kraume, M. Detailed numerical simulations of catalytic fixed-bed reactors: Heterogeneous dry reforming of methane. *Chem. Eng. Sci.* **2015**, *122*, 197–209. [CrossRef]

26. Zhang, M.H.; Dong, H.; Geng, Z.F. Computational study of flow and heat transfer in fixed beds with cylindrical particles for low tube to particle diameter ratios. *Chem. Eng. Res. Des.* **2018**, *132*, 149–161. [CrossRef]

27. Brackbill, J.U.; Kothe, D.B.; Zemach, C. A continuum method for modeling surface tension. *J. Comput. Phys.* **1992**, *100*, 335–354. [CrossRef]

28. CD-Adapco, STAR-CCM+; CD-adapco: Munich, Germany, 2017.

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