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Multiphase Catalytic Reactors: a Modular Approach

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Currently, state-of-the-art approaches to simulating the behaviour of trickle-bed reactors (TBRs) have focused solely on methods requiring high computational time and are unable to tackle systems with a large number of particles. In this work, a modular methodology based on a Lagrangian approach to TBR modelling is presented, which overcomes these drawbacks by implementing a simulation framework where different modules are interconnected and relevant information is transferred between them. The novelty of this framework stems from its adaptable configuration and its modular and unified setup, enabling it to accommodate both local and global multiscale events. The proposed methodology includes modules for the packing generation, liquid flow simulation, and of reaction system modelling within the reactor. To illustrate the procedure, a case study is discussed while demonstrating the potential of the presented approach. The results were validated against data obtained from a purpose-built experimental setup showing good agreement. The main advantages of this approach lie in its efficiency, the interrelation between different modules, and its ability to capture a wide range of information and phenomena.

1. Introduction

Catalytic reactions are the backbone of various industrial applications, including chemical, petrochemical, and biochemical processes. Specifically, Trickle Bed Reactors (TBRs) are widely employed for a broad class of catalytic reactions, including hydrogenation, oxidation, and alkylation, and are the most commonly used type of multiphase reactors due to their large surface area, low operational cost, and minimal catalyst loss (Azarpour et al., 2021).

Despite the recent advancements in experimental techniques and computational tools, understanding the microscopic and macroscopic phenomena during TBR operation still poses significant challenges. Factors such as catalyst bed specifications (e.g., particle size and bed porosity), flow distribution inside the reactor, catalyst wetting characteristics, various interphase and interparticle transport, reaction phenomena, as well as reaction kinetics, increase the complexity of rigorous TBR modelling (Srivastava et al., 2023). In the literature, empirical models or Computational Fluid Dynamics (CFD) simulations have been extensively implemented to describe the system's behaviour and performance (Jurtz et al., 2019).

Empirical correlations, derived from experimental data and observations, are used to predict various quantities based on TBR performance, such as mass and heat transfer coefficients, liquid holdup and pressure drop. However, the use of these data-based models yields reliable results only within a specific range of operating conditions that are often not valid across different reactor scales or operating conditions (Qi et al., 2021).

CFD modelling is a widely used tool for simulating and comprehending the behaviour of the fluid inside catalytic multiphase reactors. To perform a CFD simulation, the following steps are required: a) construction of the packing geometry, b) selection of the mesh method, c) definition of boundary and initial conditions, and d) solution of the Navier - Stokes (NS) equations (Zhang et al., 2013). The main limitations of this methodology include high computational burden, numerical instability, the use of a small number of catalytic particles, and the inability to achieve two-way communication between different scales (Azarpour et al., 2021).

To conclude, state-of-the-art methods exhibit the following limitations:

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- Important interactions between the phases and catalytic particles are not taken into consideration, leading to inaccurate results.
- The local structure of the catalytic packing and parameter uncertainty are not fully considered, which affects the modelling and behaviour of TBRs.
- CFD simulations require high computational effort to solve systems with a large number of particles (>25,000 particles).

Therefore, there is a need to overcome these disadvantages, and new approaches to the modelling of TBRs should be introduced. In this work, an intuitive tool for the design and analysis of TBRs operating in the low-interaction regime based on the local structure of the packed bed is presented. The proposed algorithmic framework is modular, allowing for seamless information transfer between them. Each module can be readily expanded to incorporate more representations that are detailed.

2. Solution framework description

This section presents the solution framework for modelling the behaviour of TBR. It is organised into three sequential modules interconnected, as illustrated in Figure 1. Initially, the catalytic packing is created, defining the geometry of the reactor and the particle morphology and determining the packing structure along with its axial and radial porosity. This information is essential for the Holdup calculation module, which computes the number of contact points and the static holdup.

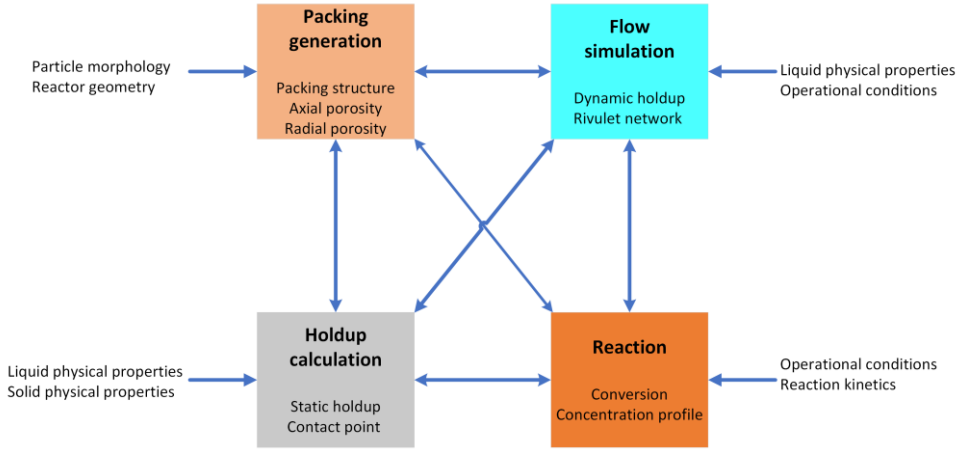

Figure 1: Schematic representation of the proposed framework

Once the bed morphology is established, the hydrodynamic (Flow simulation) module is employed to visualise flow patterns within the reactor and to compute the dynamic holdup. This calculation integrates data on the physical properties of the liquid, the bed structure from the packing generation module, and contact points. Lastly, the reaction module is engaged during catalytic reactions to analyse conversion and concentration profiles inside the TBR. This assessment is based on reaction kinetics, the rivulet network, and the holdup information. A detailed description of each module within the unified framework is given below.

2.1 Packing generation module

In the low-interaction regime, liquid maldistribution occurs when the liquid phase poorly irrigates some areas of the catalytic packing. This is one of the most significant factors in TBR operation and design (Suneja and Roy, 2023). The variation of local structural properties of the packing, such as the arrangement of particles in the catalytic bed and packing density, as well as improper initial feed distribution, liquid properties, or reactor operational conditions, can significantly influence the TBR performance. Issues such as the existence of hot spots and catalyst deactivation are common consequences (Azarpour et al., 2021). Therefore, it is of paramount importance to understand the local structural characteristics of the reactor's packing.

In this work, an optimisation approach based on the rain model is introduced. Initially, a layer of particles is placed at the bottom of the reactor, while the remaining particles are introduced and randomly placed within the reactor's space. After this initial placement, a compression and shaking algorithm based on the Monte Carlo approach is employed. In this algorithm, the particles are compressed in the z direction and adjusted at the x and y coordinates, to obtain more accurate and realistic packing structures with a lower voidage compared to existing methods. Additionally, supplementary routines are implemented to check the feasibility of the generated

catalytic bed in terms of particle placement inside the reactor, between neighbouring particles, and between particles and the wall. After constructing the packing, the proposed algorithm generates global and local geometric information, including the coordinates of each particle and the number of contact points between particles and between particles and the wall, which are available for the computations performed within the hydrodynamics module. The produced structure of the catalytic bed can be saved and reused.

2.2 Hydrodynamics module

Liquid flow distribution is a critical aspect of the design and operation of TBRs. The flow phenomena inside the reactor are challenging to describe and predict due to the random pore structure within the unit (Azarpour et al., 2021). When the liquid enters the reactor and flows through the packing, the liquid holdup can be classified as dynamic and static. In the static holdup, the liquid can be trapped between the contact points of particles, creating pendular structures and liquid pockets. In the dynamic holdup, a liquid element can appear as rivulets, films or single droplets (Srivastava et al., 2023).

In the proposed scheme, a Lagrangian approach is introduced to simulate the liquid flow distribution based on bed morphology. This method tracks individual fluid particles or elements in a three-dimensional liquid distribution network inside the packing, where discrete models represent the liquid flow as a sum of single discrete liquid elements at the particle scale (Schwidder and Schnitzlein, 2010). As the liquid elements move through the reactor's height, they can undergo different actions, such as rolling, splitting, or coalescing (Schwidder and Schnitzlein, 2012). The behaviour of each discretized liquid element is primarily influenced by the physical properties of the liquid–solid system and, to a small extent, by random actions to accurately capture the flow within the reactor’s bed morphology (Schwidder and Schnitzlein, 2012). Irregular and complex particle geometries can be handled with ease by tracking the movement and interactions of fluid elements within the void spaces between the particles of the bed. In this approach, assumptions of complete mixing in cavities and displacement in the channels connecting these cavities are applied. The dynamic holdup is obtained by summing over all the discrete liquid elements within the packing.

The same network is used to obtain the mixing characteristics of liquid flow inside the catalytic bed. Specifically, the rivulets that flow on the particle surface and the trapped liquid within the inter-particle spaces constitute the edges and vertices of the network. A Continuous-Stirred Tank Reactor (CSTR) is used to describe the vertices, while the stagnant liquid in the interparticle area is modeled as a Plug Flow Reactor (PFR), where experimental measurements are required to calculate the reactor’s characteristic parameters (Schwidder and Schnitzlein, 2012). In addition to simulating the liquid flow inside the reactor, the hydrodynamic module determines the axial and radial voidage distribution and the local degree of wetting. This representation of liquid flow as a set of CSTRs and PFRs requires less computational time compared to CFD simulations. Overall, the proposed representation provides a detailed, flexible and accurate method for modelling the complex dynamics of the TBRs, enabling better prediction and optimisation of the reactor performance.

2.3 Reaction module

One of the advantages of TBRs is their high conversion and selectivity due to low axial dispersion (Zhao et al, 2020). Therefore, it is of utmost importance to quantify and examine the performance of the catalytic reactor in the low-interaction regime in terms of the reaction system. The network of reactors produced by the hydrodynamics module, along with the knowledge of the local degree of wetting and the system’s operating conditions, form the basis for the reaction module. This module can predict the conversion inside the reactor and the residence time distribution for a given heterogeneous catalytic system through dynamic mass balances (Schwidder and Schnitzlein, 2012). Specifically, the user defines the reaction kinetic model for the examined reaction system and the corresponding mass transport parameters, which are calculated by experimental measurements. An initial demonstration and experimental validation of the proposed reaction module were conducted for the hydrogenation of alpha-methylstyrene to cumene (Schwidder and Schnitzlein, 2012).

3. Results and Discussion

In the following section, a case study is examined using the methodology proposed above while introducing the implementation of the modular and Lagrangian approaches. For this case study, the following assumptions are considered:

1. The TBR operation is assumed to be isothermal at 25 °C.
2. The catalytic reactor is operated at atmospheric pressure (1 atm).
3. Water is considered as the liquid phase.
4. The shape of the catalytic particles is assumed to be spherical with constant diameter.
5. The pressure drop along the length of the reactor can be neglected.
6. The presence of reaction is not taken into consideration.

Table 1 displays the parameter values for the bed generation of a selected example scenario, along with the obtained values of the number of contact points between particles and the wall–particle interface. Additionally, the axial and radial distributions of the voidage and their mean values are presented in Figure 2.

Table 1: Packing characteristics

Description	Value	Units
Reactor diameter	0.1	cm
Reactor length	0.61	m
Number of particles	26,511	-
Particle diameter	6	mm
Particle – particle contact points	108,646	-
Wall – particle contact points	4,842	-
Total static holdup	0.023067	-

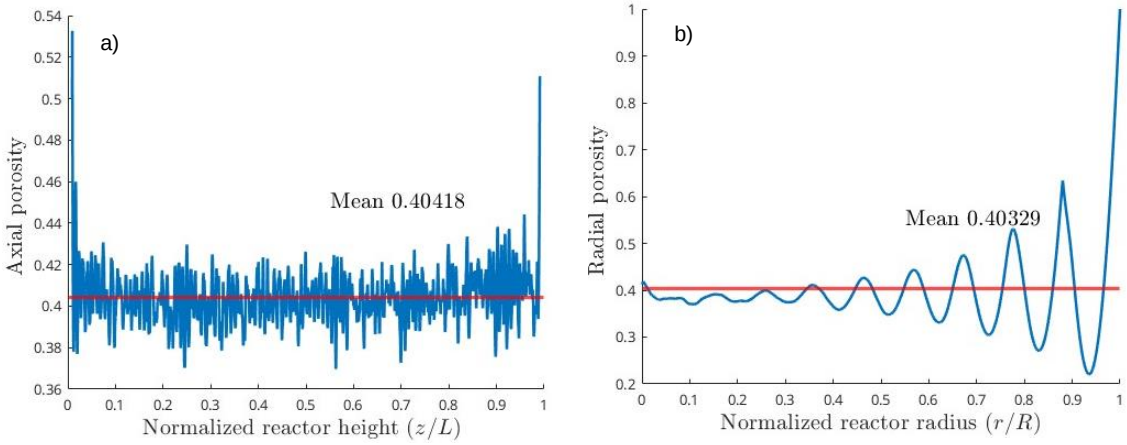

Figure 2: a) Axial and b) radial porosity distribution

The structure of the catalytic bed is subsequently transferred to the holdup module, where the holdup distribution is obtained. It is evident from Figure 3a that the static holdup near the reactor's wall has the highest value along the axial direction. This behaviour can be explained by exclusively considering the contact points formed between particles and the wall in the static holdup calculation. The proposed methodology also enables the radial distribution generation of the contact points inside the catalytic bed. Figure 3b illustrates that the number of contact points near the wall is lower in comparison to those in the interior of the packing. This difference in the distribution of contact points can be justified by the fact that two spheres near the wall form a contact point at a greater distance in contrast to the contact points in the interior of the reactor.

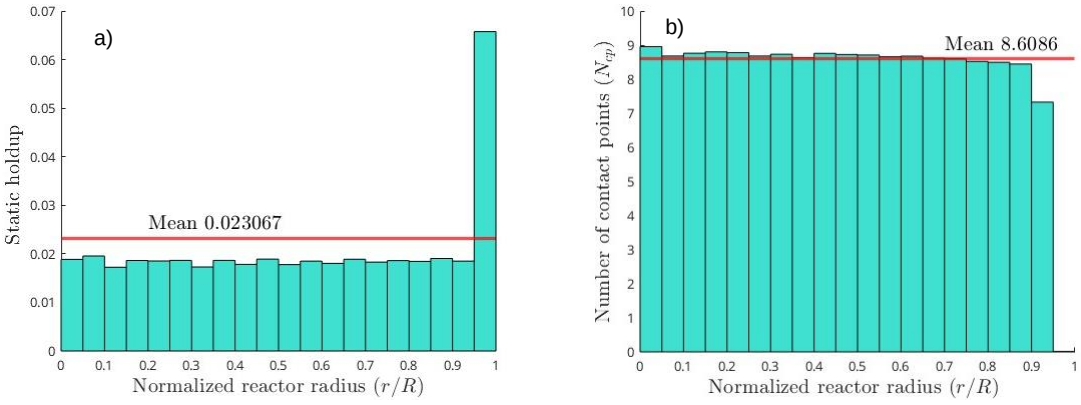

Figure 3 a) Static holdup and b) distribution of the number of contact points

After obtaining and transferring the information from the packing and holdup modules, the hydrodynamic module is used to determine the liquid flow distribution. In this case, the liquid flow rate entering the catalytic reactor is set to 250, 525, and 780 mL/min, and it enters from the top side of the reactor. The hydrodynamic module successfully calculates the number of rivulets created inside the reactor, the total wetting of the bed, and the ratio of wetted spheres for all the liquid flow values, as shown in Table 2.

Table 2: Flow distribution characteristics

Description	Value			Units
Liquid flowrate	250	525	780	mL/min
Number of rivulets	8,756	12,366	13,347	-
Wetting	0.0370	0.0506	0.0570	-
Wetted spheres	0.1288	0.1696	0.1809	-

Wetting efficiency is a crucial aspect in the development and operation of industrial TBRs and plays an important role in the degree of catalyst usage, mass transfer rate and possible creation of hotspots (Azapour et al., 2021). A higher number of rivulets ensures a uniform liquid distribution, enhancing mass transfer, while higher total wetting and a higher fraction of wetted spheres increase the effective catalyst surface area, improving reaction rates and heat transfer. It should be noted that the particle positions in the generated catalytic bed might not match those of an actual packing. Therefore, the liquid flow distribution obtained from the solution algorithm may not necessarily agree with experimental data.

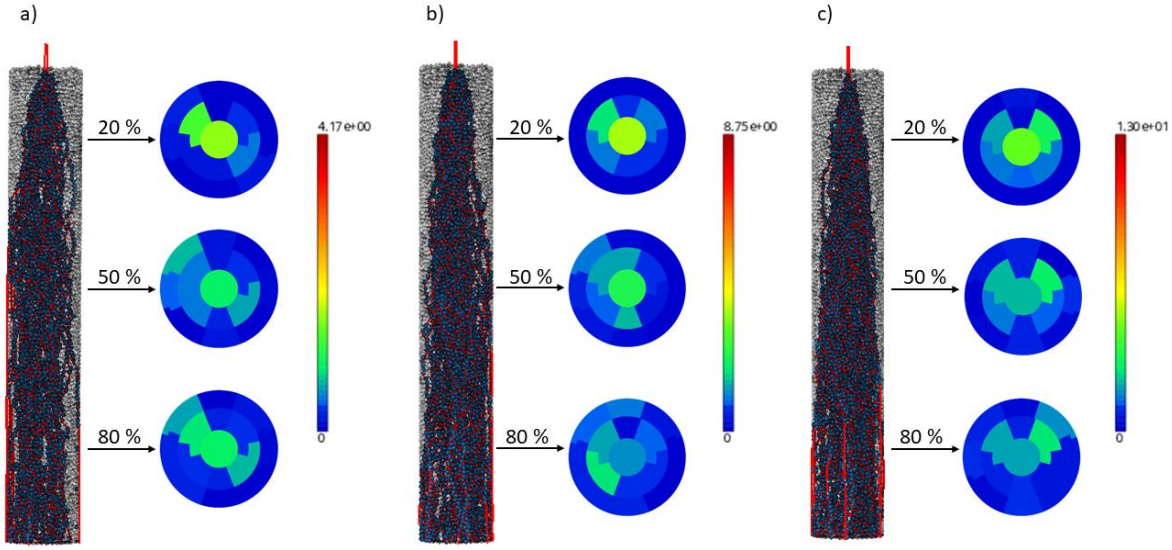

Figure 4: Liquid flow distribution for a flow of a) 250 mL/min, b) 525 mL/min, and c) 780 mL/min

The simulated liquid distribution is represented as a three-dimensional graph network, and the radial flow distribution for different horizontal cuts through the reactor for the 250, 525, and 780 mL/min flow rates are shown in Figure 4. It can be observed that the flow in the centre of the catalytic reactor exhibits the highest value along the reactor's height. As the liquid flows inside the catalytic bed, the effects of branching and channelling take place, owing to the existence of gravitational, inertial, viscous, surface and interfacial forces. Particularly, the rivulets forming close to the reactor's wall appear after half of the reactor's length. The same behaviour is exhibited for all the examined liquid flow rate values. The dynamic holdup values obtained by the proposed solution framework are presented in Figure 5. It can be observed that the obtained dynamic holdup values are in good agreement with the experimental data.

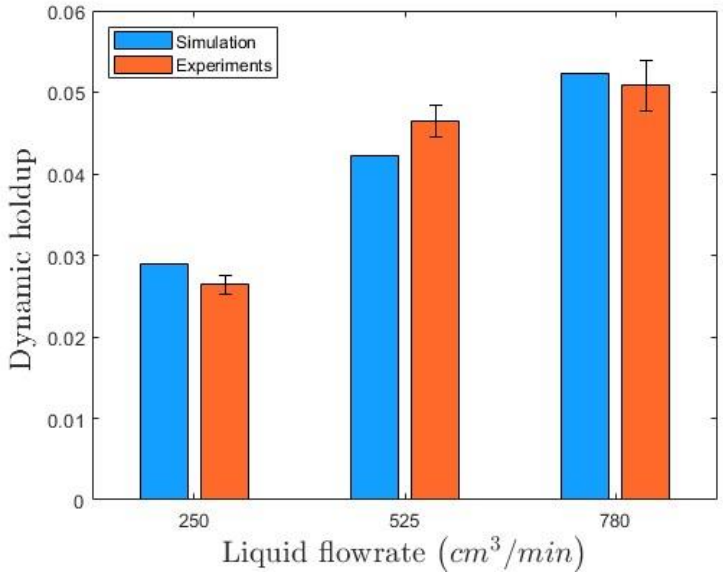

Figure 5: Dynamic holdup values for different liquid flow rates

4. Conclusions

This contribution introduces a modular and intuitive approach to designing and analyzing TBR behavior using a Lagrangian approach. The proposed solution scheme leverages both local and global characteristics of the catalytic bed, facilitating seamless information transfer between different modules. The novelty of this framework lies in its ability to account for multiscale information at both local and global levels due to its flexible and modular setup. This approach effectively captures the multiscale nature of phenomena occurring at the macro-, meso- and micro-scale of the reactor. Numerical results demonstrate the practicality and effectiveness of this methodology in handling bed morphologies with a large number of particles (>25,000 particles), consequently, delivering results within reasonable timeframes. This is in contrast to CFD, which often struggle with multiscale coupling and two-way communication between different scales, and is computationally expensive. Furthermore, this methodology holds the potential for extension to various types of particle geometries (e.g., cylinders, Raschig rings, trilobes, etc.) and can accommodate the inclusion of the gas phase or considerations related to the pressure drop inside the reactor.

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