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CFD Modelling of Trickle-Bed Reactors for Wastewater Treatment

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Abstract

This study aims to incorporate most recent multiphase models in order to investigate the hydrodynamic behaviour of a TBR in terms of pressure drop, liquid holdup and catalyst wetting efficiency. Taking into account transport phenomena such as mass and heat transfer, an Eulerian *k*-fluid model was developed resulting from the volume averaging of the continuity and momentum equations and solved for a 2D representation of the bed at unsteady state. CFD model predicts hydrodynamic parameters with reaction conditions quite well. Moreover, catalytic performance is investigated in terms of TOC and one can conclude that Eulerian *k*-fluid model is a rational choice for flow simulation in packed beds, if good closures for fluid/fluid and fluid/particle interactions are incorporated in the CFD model used. The CFD approach can investigate external catalyst wetting in downflow flow mode performing a benchmark of hydrodynamic parameters encountered in the commercial-scale TBR deployment for CWAO technology in advanced wastewater treatment facilities.

Keywords: CFD, multiphase flow, hydrodynamics, environmental pollution

1. Introduction

Major advances for the chemical industries will, no doubt, continue to emerge from catalysis, chemistry and systems engineering. However, maximizing the

industrial benefit from these fields require comparable advances in the design of chemical process equipment. Efficient and effective design ensures the delivery of materials and energy at the right places and at the right times by manipulating underlying fluid dynamics.
Our aim encompasses an upgrade of multiphase fluid modelling to allow process engineers to predict and manipulate the desired fluid dynamics in wastewater process equipment. In this paper, we provide our perspective of the current status and potential of CFD for designing process equipment with emerging multiphase flows models and CFD code approaching for computational flow modelling in wastewater treatment plants, namely in trickle-bed reactors (TBR) operation. The open literature is reviewed and an Eulerian-Eulerian multidimensional model for the TBR unit is presented.

2. Problem Statement, background

An emerging area in lifecycle environmental applications involves multiphase flows in advanced processes for wastewater treatment, which represents a great challenge for aquatic flora and fauna preservation. In fact, bioremediation technologies have known limitations and alternative destruction methods such as catalytic wet air oxidation (CWAO) have been conducted on a variety of organic compounds using numerous catalysts with a great potential in advanced wastewater treatment facilities [1].
Trickle-bed reactors (TBR) are widely used for heterogeneous catalyzed reactions between gas and liquid reactants, such as hydrogenation, oxidation or partial oxidation and detoxification of liquid effluents. In these reactors, gas and liquid phase flow co-currently downward through a fixed bed of catalyst particles. Though most of the research studies before 1990 have been performed at atmospheric pressure, a considerable number of investigations were undertaken in pressurised trickle-bed reactors after that period [2]. A large number of studies have been reported in the literature on the various hydrodynamic aspects of TBRs remaining a lot of work to be done in environmental reaction engineering. Meanwhile, relatively few investigations have been published concerning catalytic liquid-phase oxidation of organic compounds in large-scale TBRs for wastewater treatment where hydrodynamic parameters prevail. In the present paper, TBR was modelled to bring up for the first time hydrodynamic and reactions studies by means of CFD codes making possible the investigation of the efficiency for different hydrodynamic regimes involved in continuous operation of TBR.

3. Paper approach

TBR models reported in the literature considered isothermal operation and use a pseudo-homogeneous approach or heterogeneous model with plug-flow for gas and liquid phase with some models accounting for liquid flow non-uniformity

and maldistribution by using an axial dispersion model [2,3]. To improve the capability of multiphase reactor models, one has to solve the complete multidimensional flow equations coupled with chemical species transport, reaction kinetics, and kinetics of phase change. An Eulerian model was proposed by the authors for the prediction of hydrodinamic behaviour.

Methodology and case study

In the Euler-Euler concept, sometimes called the two-fluid approach, both the continuous and dispersed phases are considered as continuous media. These models incorporate two-way coupling, which is especially important for high voidage flows. The interfacial momentum transfer between the liquid and the gas includes a number of force contributions, form and viscous drag, mass force which is an inertial force caused by relative acceleration, the effect of turbulent fluctuations on the effective momentum transfer, and the lift force which denotes the transverse force caused by rotational strain, velocity gradients, or the presence of walls. The CFD model equations were implemented in commercial software FLUENT (of Fluent Inc., USA) using user defined routines. Only the main conservation equations are presented. The continuity and momentum balance for phase *q* are written in Eq. (1) and (2), respectively:

$$\frac{\partial}{\partial t}(\alpha_q \rho_q) + \nabla \cdot (\alpha_q \rho_q \vec{v}_q) = \sum_{p=1}^{n} (\dot{m}_{pq} - \dot{m}_{pq}) + S_q \tag{1}$$

$$\frac{\partial}{\partial t}(\alpha_q \rho_q \vec{v}_q) + \nabla \cdot (\alpha_q \rho_q \vec{v}_q \vec{v}_q) = -\alpha_q \nabla p + \nabla \cdot \overline{\overline{\tau}}_q + \alpha_q \rho_q \vec{g} + \sum_{p=1}^{n} (\vec{R}_{pq} + \dot{m}_{pq} \vec{v}_{pq} - \dot{m}_{qp} \vec{v}_{qp}) + (\vec{F}_q + \vec{F}_{lift,q} + \vec{F}_{vm,q}) \tag{2}$$

Eq. (2) must be closed with appropriate expressions for the interphase force $\vec{R}_{pq}$. This force depends on the friction, pressure, cohesion, and other effects, and is subject to the conditions that $\vec{R}_{pq} = -\vec{R}_{qp}$ and $\vec{R}_{qq} = 0$. The form of a simple phase interaction term is given by Eq. (3):

$$\sum_{p=1}^{n} \vec{R}_{pq} = \sum_{p=1}^{n} K_{pq} (\vec{v}_p - \vec{v}_q) \tag{3}$$

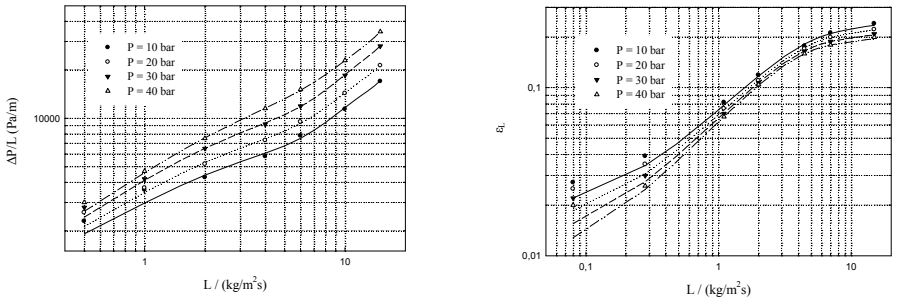
where $K_{pq} = K_{qp}$ is the interphase momentum exchange coefficient. To describe the conservation of energy in Eulerian multiphase applications, a separate enthalpy equation should be written for each phase as shown in Eq. (4):

$$\frac{\partial}{\partial t}(\alpha_q \rho_q h_q) + \nabla \cdot (\alpha_q \rho_q \vec{u}_q h_q) = \\ -\alpha_q \frac{\partial p_q}{\partial t} + \overline{\overline{\tau}}_q : \nabla \vec{u}_q - \nabla \cdot \vec{q}_q + S_q + \sum_{p=1}^{n} (\vec{Q}_{pq} + \dot{m}_{pq} h_{pq} - \dot{m}_{qp} h_{qp}) \quad (4)$$

Turbulent flows are characterized by fluctuating velocity fields mixing transported quantities such as momentum, energy, and species concentration, and cause the transported quantities to fluctuate as well. It was used the *k-ε* model which is a semi-empirical model based on model transport equations for the turbulence kinetic energy (*k*) and its dissipation rate (*ε*). CFD studies were performed by taking into account the geometry of a TBR pilot plant unit installation at our laboratory. The cylindrical reactor was made of stainless steel (SS-316) with 50 mm of internal diameter and 1.0 m length according to the reactor unit used by Levec and coworkers [4,5]. The boundary conditions of the mesh were designed in a CAD commercial program (GAMBIT).

Results & discussions

The mesh was validated by checking the mesh sensitivity and by comparing the numerical results against the single-phase and two-phase experimental data available from the work developed by Levec and coworkers [4,5]. The mesh adopted in the TBR reactor is tetrahedral around and over the catalyst particles and hexahedral elsewhere. The cell number is around 800,000. The momentum equations are solved with the coupling SIMPLE algorithm and the second upwind discretization scheme.

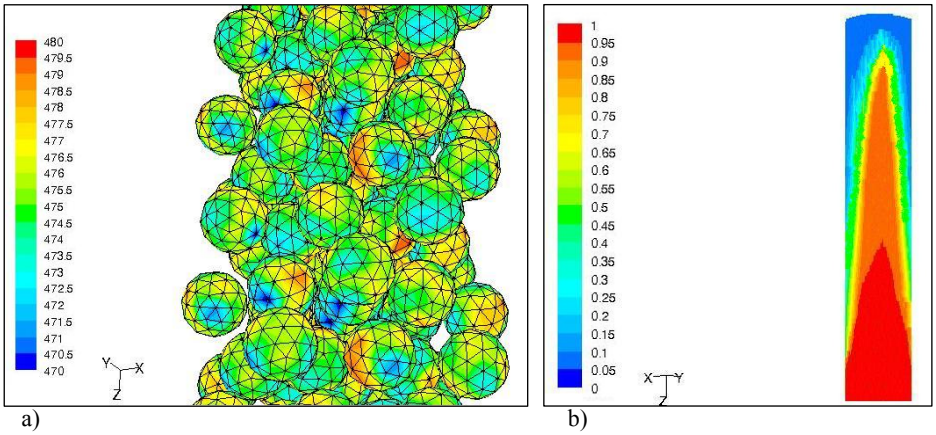


a) b)

Fig. 1: *(a)* Pressure drop and *(b)* liquid holdup as a function of liquid mass flux at different operating pressure (curves: CFD results; points: experimental values from the literature [4,5])

The pressure is computed by means of the PRESTO scheme. In Fig. 1a) and b), pressure drop and liquid holdup modelling results for different operating pressures are compared with experimental data taken from the open literature

[4,5]. The CFD model fitted very well pressure drop and liquid holdup pointing out that when pressure ranges from low values, P = 10 bar, to higher values, P = 40 bar, liquid holdup decreases in accordance with Fig.1b). One can then conclude that the hydrodynamic behaviour in terms of liquid holdup and pressure drop seems to be well described by the Eulerian model, reflected by the fact that all the data lay on an expected performance for the packed bed. In order to investigate the oxidation process in TBR, CFD simulations were performed using the kinetic laws previously obtained [6] for a phenolic acids mixture containing syringic, vannilic, *3,4,5*-trimethoxybenzoic, veratric, protocatechuic and *trans*-cinnamic acids present in wastewaters from olive oil mills. In the CFD model it was assumed that chemical reaction occurs in catalyst pores and at the catalyst surface. External mass transfer limitations were taken into account in order to guarantee that the process is not only governed by the chemical reaction, but assumes also the competition between transport and reaction that it is present in the operation of large scale pilot plant units. Runs were performed in unsteady state mode to investigate the unit start-up and to evaluate the TBR performance until steady state was reached. Our reaction studies indicated that steady state of TBR unit is achieved practically in 2 *h* and catalyst surface temperature colour map taken at this time is shown in Fig. 2a). The different temperatures attained in different locations of the catalyst particles indicate different reaction rates (for the exothermic oxidation process of the pollutants), which reflect different wetting levels of the solid by the liquid effluent.



a) b)

Fig. 2: *(a)* Catalyst surface temperature (*K*) and *(b)* TOC degradation colour map at *t = 2 h*

Hence, liquid holdup plays an important role in TBR hydrodynamics that affects the catalyst wetting efficiency, which, in turn, affects the reaction selectivity depending on whether the reaction takes place solely on the wetted catalyst area or on dry and wetted catalyst areas alike. According to Fig. 2a), it

is possible to identify this fact in terms of catalyst surface temperature with values charged from 470 K to 480 K. As one can observed in Fig. 2b), the corresponding TOC (Total Organic Carbon) profiles bring up that catalyst wetting is also a performance rating parameter in the degree of packed bed utilization. Therefore, the evaluation of external catalyst wetting efficiency is an important design and scale-up parameter in determining the degree of catalyst utilization in trickle-bed reactors where the internal contacting is usually equal to unity due to capillary effects. CFD models bring usefulness at this point with the sacrifice of higher computational power .

4. Conclusions

Eulerian multiphase CFD model has made great progress in order to investigate the phases interaction coupled with turbulence models. CFD Eulerian model predicts reasonably well hydrodynamic parameters with reaction conditions and catalytic performance is investigated in terms of TOC. Eulerian *k*-fluid model is a rational choice for flow simulation in packed beds but in order to achieve feasibility of continuous CWAO technology in wastewater treatment, any advance in TBR technology will thus represent substantial savings, and this stimulates the continued research efforts aimed at improving TBR operation and performance by means of CFD codes.

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References

1. S. K. Bhargava, J. Tardio, J. Prasad, K. Foger, D. B. Akolekar and S. C Grocott, Ind. Eng. Chem. Res., 45 (2006) 1221.
2. M. P. Dudukovic, F. Larachi, P. L. Mills, Catal. Rev., 1 (2002) 123.
3. P. R. Gunjal, M. N. Kashid, V. V. Ranade, R. V. Chaudhari, Ind. Eng. Chem. Res., 44 (2005) 6278.
4. A. Lakota, J. Levec, R. G. Carbonell, AIChE J., 48 (2002) 731
5. D. Nemec, J. Levec, Chem. Eng. Sci., 60 (2005) 6958.
6. R. J. G Lopes, A. M. T. Silva, R. M. Quinta-Ferreira, Appl. Cat. B (2006), *in press*