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CFD of Multiphase Flow in High pressure Trickle Bed Reactors: Porous Media Concept

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Abstract

A two-phase Eulerian model, based on porous media concept, describing the flow domain as porous region is presented to estimate the hydrodynamics of two-phase flow in trickle-bed reactors (TBRs) operating at high pressures. The drag forces between phases have been accounted by employing the relative permeability concept (Sàez and Carbonell, 1985). The model has been validated with the different sets experimental data obtained from different independent sources. All the comparisons lead to the fact that the model functions arguably well in predicting the experimental data for high pressure operations. The developed model is very much flexible unlike the traditional CFD approach, i.e. three-phase Eulerian simulations for different particle size effect incorporation without much complexity. While simulating for high pressure condition, we have applied the recently developed correlations (Nemec and Levec, 2005) for relative permeabilities.

Keywords: trickle bed reactor, CFD, porous media, two-phase flow, high pressure

1. Introduction

Trickle bed reactors are packed beds of catalyst with cocurrent flow of gas and liquid reactants. They represent an important class of multiphase reactors for carrying out gas-liquid reactions in the presence of heterogeneous solid catalyst. There are many applications of trickle bed reactors in the petroleum industry, as well as in the chemical process industry. The most extensive and popular uses of these reactors are in the field of petroleum refining industry, more specifically in hydroprocessing of oils (e.g. hydrotreating, hydrocracking). Therefore, they are also playing a major role in containment of environmental pollution.

With current interest in technologies of ‘deep’ processing, such as deep hydrodesulfurization, for achieving transportation fuel with ultra level sulfur (~10 ppm /zero ppm) due to stringent environmental legislations, the need to be able to improve the performance of the reactor is even more important. Again, the

performance of the reactor largely depends on combination of catalyst effectiveness, proper reactor design and appropriate process conditions (Nigam and Larachi, 2005).

For conventional desulfurization reactions, the practitioners used to encounter a severe problem of deceleration of the reaction along the reactor length due to diminution hydrogen partial pressure as it is being consumed continuously. Again, high temperature thermodynamically favors the desulfurization reactions conducted in TBRs, which consequences the gas phase expansion and in turn impedes the gaseous reactant from dissolving sufficiently into the liquid phase. To avoid these kinds of difficulties, in reality i.e. in refining industry, trickle-bed reactors are generally operated at high pressures up to about 20–30MPa in order to slow down catalyst deactivation, to improve the solubility of the gaseous reactant and therefore to attain high conversion (Attou, 1999).

While scale-up of trickle bed reactors in petroleum processing for certain well characterized crude oil feedstock is probably well-established as a proprietary industrial art (even if based on heuristics in many cases and not totally scientifically based), *a priori* prediction of trickle bed performance or scale-up from laboratory reactors for new feedstock is still considered very risky and therefore not considered reliable for design. One reason for this is that even after decades of research efforts, transport processes in trickle-beds are not completely understood and are not readily quantified. Also, there may be strong couplings of reaction kinetics with hydrodynamics which can make dynamics of the trickle bed highly involved and non-linear. Therefore, one must approach the problem of flow modeling in TBRs by evaluating the hydrodynamic parameters at first and then coupling those parameters with reaction kinetics for overall commercial TBR design.

Pressure drop and liquid holdup are two foremost important hydrodynamic parameters to deal with as frictional pressure drop is a measure of gas-liquid and liquid-solid mass transfer (Gianetto et al., 1978) and liquid holdup is related to other important parameters, namely, pressure gradient, gas-liquid interfacial area, the mean residence time of the liquid phase, catalyst loading per unit volume, axial dispersion coefficient, and mass-transfer and heat-transfer coefficients (Attou, 1999; Narasimhan et al. 2002). There are numerous models available for predicting these hydrodynamic parameters in the open literatures which can be broadly classified into two different categories on the basis of *empirical* approach and *theoretical* or *semi-empirical* approach (details in Atta et al. 2007a and references therein). Most importantly, it can be noted that in maximum cases, the studies have been limited to the atmospheric pressure condition. Surprisingly, there is rather scarce amount of research dealing with investigation of pressure drop and liquid saturation in TBRs operating at high pressures (Wammes and Westerterp, 1990; Wammes et al., 1990 and 1991; Larachi et al. 1991; Al-Dahhan and Dudukovic, 1994; Nemec et al., 2001; Narsimhan et al. 2002; Nemec, 2003; Nemec and Levec, 2005). In addition to this, despite increasing computational power and development of Computational Fluid Dynamics (CFD) and its extensive applications to the modeling of multiphase flow in TBR (Jiang et al., 2002a, b; Gunjal et al., 2003 and 2005; Atta et al, 2007a, b), to the best of our knowledge, a CFD based model to predict the hydrodynamics of two phase flow in TBR operating at high pressure is yet to be established.

Therefore, with the current interest in numerical prediction of hydrodynamics of trickle bed reactors operating at high pressures, in this present article we propose to use our previously established CFD model (Atta et al., 2007a) with the incorporation of modified relative permeability correlations which are developed very recently by Nemec and Levec (2005) for TBR operating at high pressure (even up to 50 bar). The independent experimental data sets reported by Al-Dahhan and Dudukovic, 1994 were selected in the present work to validate the predictions.

2. Modeling

It can be observed that most of the literature available dealing with trickle bed flow simulation uses a three-phase Eulerian model in which the solids velocity is identically set to zero. The fluid phases (gas and liquid) are allowed to flow freely through this *bed* of solids as per the conservation equations and closure models. Such a calculation is computationally demanding and yet very difficult to implement in the case of predicting hydrodynamics of high pressure TBRs. In this research work, we extend the use of previously established CFD model (Atta et al., 2007a) for the estimation of pressure drop and liquid hold-up in trickle-bed reactors operating at high pressures by implementing the concept of porous media with less computational effort. Based on porous media concept, a two-phase Eulerian model describing the flow domain as porous region has been presented to forecast the hydrodynamics of two-phase flow in trickle-bed reactors (TBRs) under cold-flow conditions using FLUENT 6.2 software (of Fluent. Inc., USA). The drag forces between phases have been accounted by employing the relative permeability concept (Sàez and Carbonell, 1985). However, the correlations for relative permeabilities (of liquid and gas phases) have been adopted from Nemec and Levec (2005).

The model equations describing the gas and liquid two phase flow through the packed bed are based on phasic volume fraction concept. Volume fractions represent the space occupied by each phase, and the laws of conservation of mass and momentum are satisfied by each phase individually.

The volume-averaged equations for each flowing phase can be written as:

- Continuity equation

$$\frac{\partial(\varepsilon\rho_\alpha s_\alpha)}{\partial t} + \nabla.(\rho_\alpha u_\alpha) = 0\,,\ \alpha = g,l \tag{1}$$

- Momentum balance equation

$$\rho_\alpha\left(\frac{\partial u_\alpha}{\partial t} + u_\alpha.\nabla u_\alpha\right) = -(\nabla p_\alpha - \rho_\alpha g) + \nabla.(\tau_\alpha + R_\alpha) + F_\alpha \tag{2}$$

where F_α = Total drag force per unit of bed volume exerted by the phase α

τ_α and R_α are, respectively, the volume averaged viscous stress tensor and the turbulence stress tensor of phase α. Inter-phase coupling terms accounted by equation (2) are based on relative permeability concept developed by Sáez and Carbonell (1985) which states that:

$$\frac{F_\alpha}{\varepsilon_\alpha} = \frac{1}{k_\alpha}\left[A\frac{Re_\alpha}{Ga_\alpha} + B\frac{Re_\alpha^2}{Ga_\alpha} \right]\rho_\alpha g \tag{3}$$

where, A and B in the equation (3) are the Ergun equation coefficients for single-phase flow in the packed bed (Ergun, 1952). The Reynolds and Galileo numbers are defined as:

$$\mathrm{Re}_\alpha = \frac{\rho_\alpha u_\alpha d_e}{\mu_\alpha(1-\varepsilon)}$$

$$Ga_\alpha = \frac{\rho_\alpha^2 g d_e^3 \varepsilon^3}{\mu_\alpha^2(1-\varepsilon)^3} \tag{4}$$

where

$$d_e = \frac{6V_p}{A_p}$$

While evaluating for relative permeabilities, Nemec and Levec (2005) has come up with an excellent article. They have studied these parameters with 1300 newly measured data pairs of pressure drop and liquid holdup obtained for a wide range of commercially relevant operating conditions (including pressures up to 50 bar) as well as types of packing (both in terms of size and shape). Regarding the dependency as well as sensitivity of relative permeabilities on different possible parameters, they have shown by their extensive experimentation and analysis for a wide rage of operating conditions and the typical shapes and sizes of particles encountered in commercial trickle-bed reactors that relative permeabilities are solely the functions of the corresponding phase saturation. Before concluding this remark, they have carefully explored the effects of uncertainties associated with the phase relative permeabilities and also have carried out the detailed study on the phenomenological insights of the suitable correlations, e.g. the effect of particle shape & size, effect of flow rate and reactor pressure. Surprisingly, they have opposed the observation by Lakota et al. (2002) on the particle shape dependency of the gas phase relative permeability. They have argued that the effect of shape factor is accounted by Ergun constants however the relative permeability being the ratio between single and two-phase pressure drop, this shape effect has been already taken care in that respect. In this context, it is perhaps worth mentioning that the fluid–fluid interaction model (developed theoretically by Attou et al., 1999) has a notable feature of not having any adjustable parameter in the closure but still predicts accurate results when incorporated into the CFD framework (Jiang et al., 2002a, b; Gunjal et al., 2003 and 2005). This aspect leads to the drawback of complicated incorporation different particle size and shape effect in that model. Hence, the use of this model poses an

added advantage of being flexible for different particle sizes which can be incorporated without much complexity.

According to Nemec and Levec (2005), the empirically derived correlations for relative permeabilities are:

<u>liquid phase</u>:

$$k_\beta = \delta_\beta^{2.9}, \left(\delta_\beta \geq 0.3\right)$$

$$k_\beta = 0.40\delta_\beta^{2.1}, \left(\delta_\beta < 0.3\right)$$

<u>gas phase</u>:

$$k_\gamma = 0.50 s_\gamma^{3.9} \qquad (5)$$

where $s_g = 1 - \frac{\varepsilon_l^0}{\varepsilon}$ and $\delta_l = \frac{\varepsilon_l - \varepsilon_l^0}{\varepsilon - \varepsilon_l^0}$ = ratio of effective volume of flow of the liquid phase to the available volume of flow

The static liquid holdup (ε_l^0) can be calculated by the following correlation given by Sáez and Carbonell (1985). More detailed derivation and discussions of governing equations can be found elsewhere (Sàez and Carbonell, 1985; Atta et al., 2007).

3. Model Setup and Numerical Solution procedure

Considering a two-dimensional axisymmetric domain, the above set of model equations was solved using commercial software FLUENT 6.2 (of ANSYS. Inc., USA) defining the solution domain as porous. The bed dimensions were chosen according to the geometry prescribed by Al-Dahhan and Dudukovic (1994).

In order to solve the governing equations, the assumptions made in this model are:

1. There is no inter-phase mass transfer
2. The porous medium is taken to be isotropic i.e. permeabilities are independent of direction
3. The porosity is uniform and constant
4. The contribution of the turbulent stress terms to overall momentum balance equation (2) is not significant. This assumption has also been used by other authors (e.g. Jiang et al., 2002a).

Experiments with prewetted and non-prewetted bed (Jiang et al., 2001) also indicate that the macro-scale effects of capillary pressure are negligible when the particles are completely wetted.

The gas phase was treated as primary phase and liquid phase was considered as secondary phase. At the inlet, flat velocity profile for gas and liquid phases was

assumed and implemented. No slip boundary condition was set for all the impermeable reactor walls. At the bottom of the column, an outlet boundary condition was specified. Unsteady state simulations were carried out with the time step of 0.005 s. Some preliminary numerical experiments were carried out to identify the required number of computational cells to obtain grid independent results. It was also ensured with the preliminary numerical experiments to have discretization scheme independent results. These simulations confirmed that the grid size taken was satisfactory, as further increase in number of grids did not appreciably affect the predicted results. Furthermore, the numerical computation was assumed to be converged by checking mass residual (less than 10^{-4}) at different plane along the length.

4. Results and Discussions

The simulated results (considering Ergun constants A=180 and B=1.8) were validated against various published experimental data of pressure drop and liquid hold-up. However, in this communication, a few of them (in particular with Al-Dahhan and Dudukovic, 1994) have been presented. The details of operating conditions for the experimental dataset are adopted from Al-Dahhan and Dudukovic (1994). For example:

- Reactor pressure, MPa — $0.3 \leq P \leq 5.0$
- Temperature, K — 298
- System
  - Liquid Phase — Water, Hexane
  - Gas Phase — Nitrogen, Helium
- Packing
  - silica shell (sphere) — *porous*, d_p=1.52 mm, ε=0.412
  - glass beads (sphere) — *nonporous*, d_p=1.14 mm, ε=0.392
  - 0.5% Pd/alumina (extrudate) — *porous*, $(d_p)_{eq}$=1.99 mm, ε=0.355

Figures 1 and 2 show the comparison of simulated results with the experimental data of Al-Dahhan and Dudukovic (1994) for observed pressure drop per unit length and liquid holdup. For the given system of hexane and nitrogen with porous spherical silica shell, Figs. 1 and 2 represent the comparative studies of effect of reactor pressure for two different operating pressures (0.31 MPa and 3.55 MPa) on pressure drop and liquid holdup with literature data for a constant gas velocity of 8.5 cm s^{-1}. Similarly those figures also depict the comparison of effect of gas flow rate (1 cm s^{-1} and 8.5 cm s^{-1}) for a constant operating pressure of 3.55 MPa. It can clearly be observed that for a particular gas velocity, with the increase in operating pressure, frictional pressure drop per unit length increases whereas corresponding liquid holdup decreases. Again, for a constant operating pressure, increasing gas velocities result in

higher pressure drop and lower liquid holdup. All these predicted results are in good agreement with the experimental observations and facts.

We have further tested this CFD model for a different set of system (water - nitrogen) with different shape of catalyst particle (porous extrudates) to study the effect of reactor pressure on hydrodynamic parameters (pressure drop and liquid holdup). Fig. 3 and 4 exhibit the comparison of predicted results with the experimental data from open literature for 3 different operating pressures (0.31, 1.82 and 3.55 MPa) with constant gas velocity (≈ 8.5 cm s^{-1}).

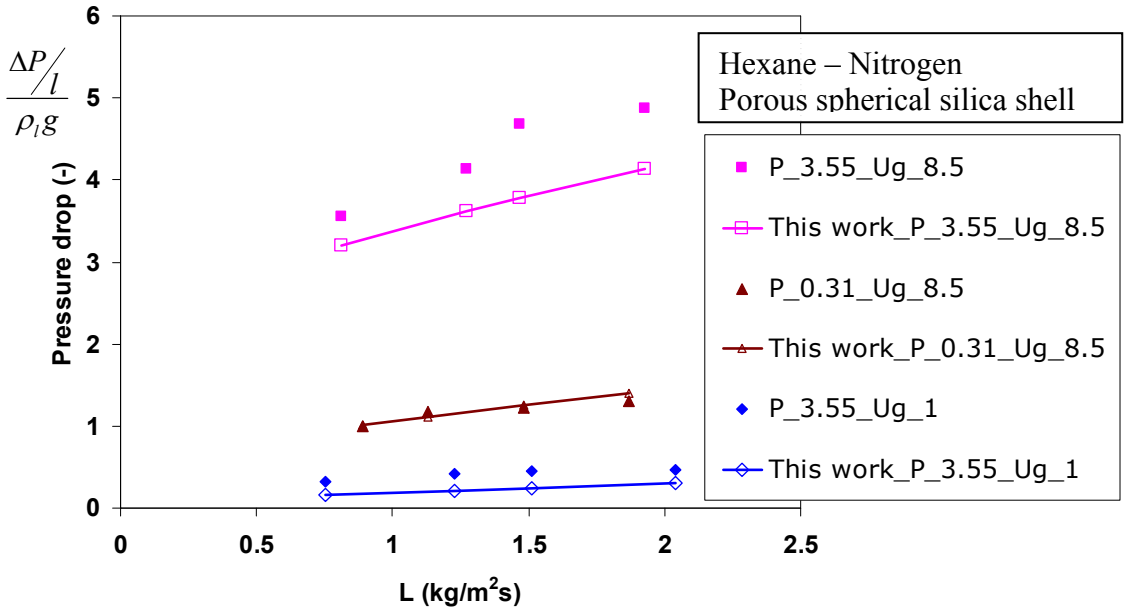

Fig 1: Comparative plot of effect of reactor pressure & gas flow rate on pressure drop with Al-Dahhan and Dudukovic (1994)

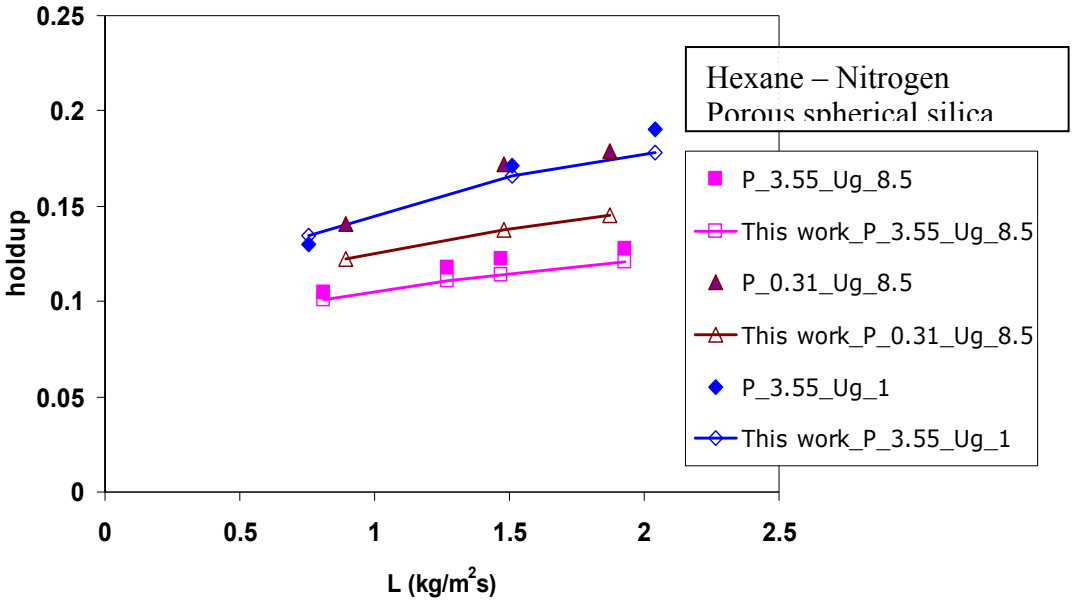

Fig 2: Comparative plot of effect of reactor pressure & gas flow rate on liquid holdup with Al-Dahhan and Dudukovic (1994)

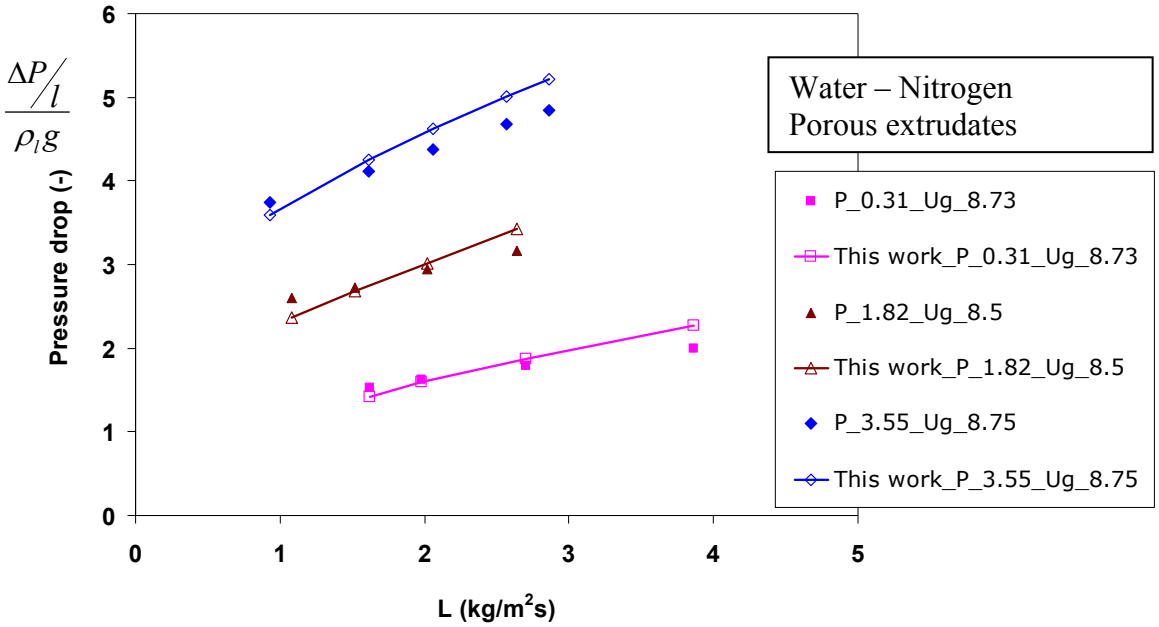

Fig 3: Comparative study of effect of reactor pressure on pressure drop with Al-Dahhan and Dudukovic (1994)

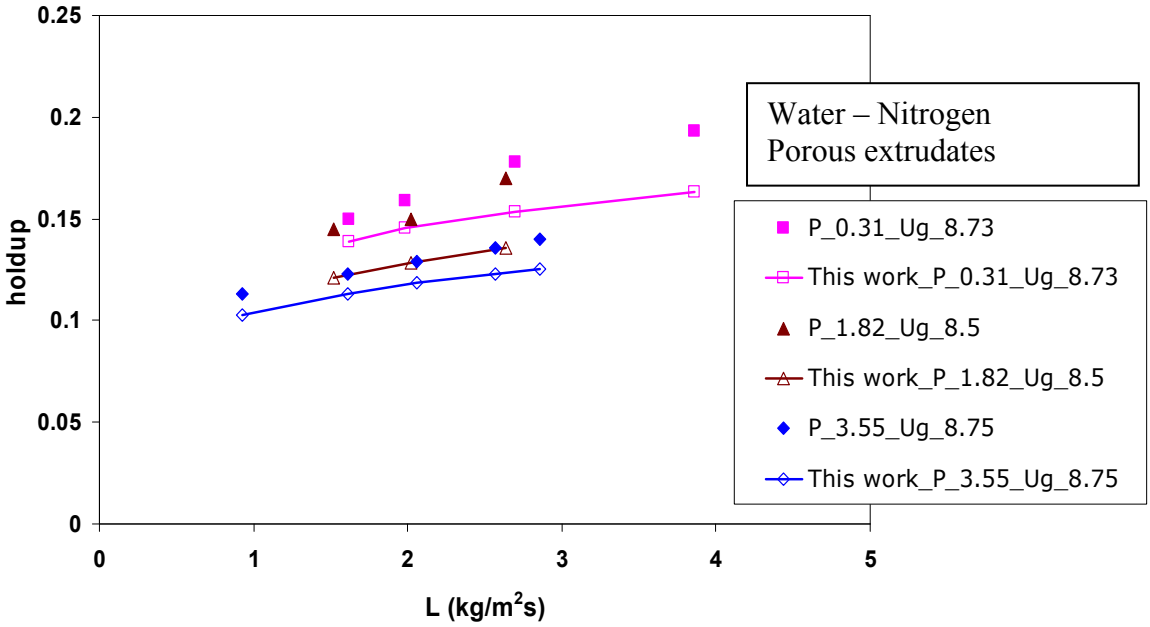

Fig 4: Comparative study of effect of reactor pressure on liquid holdup with Al-Dahhan and Dudukovic (1994)

Figures 5 and 6 represent the comparison of this model prediction with experimental data from Al-Dahhan and Dudukovic (1994) to observe the effect of different gas densities for a given system of hexane and nitrogen/helium with nonporous spherical glass beads as catalyst particles.

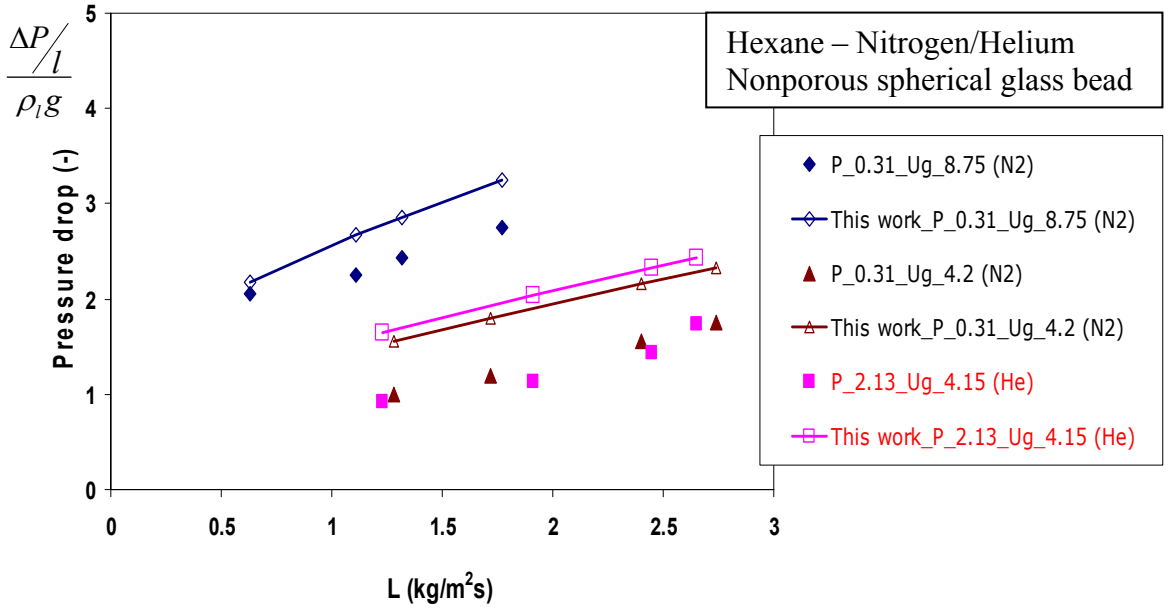

Fig 5: Comparative study of effect of gas density and flow rate on pressure drop with Al-Dahhan and Dudukovic (1994)

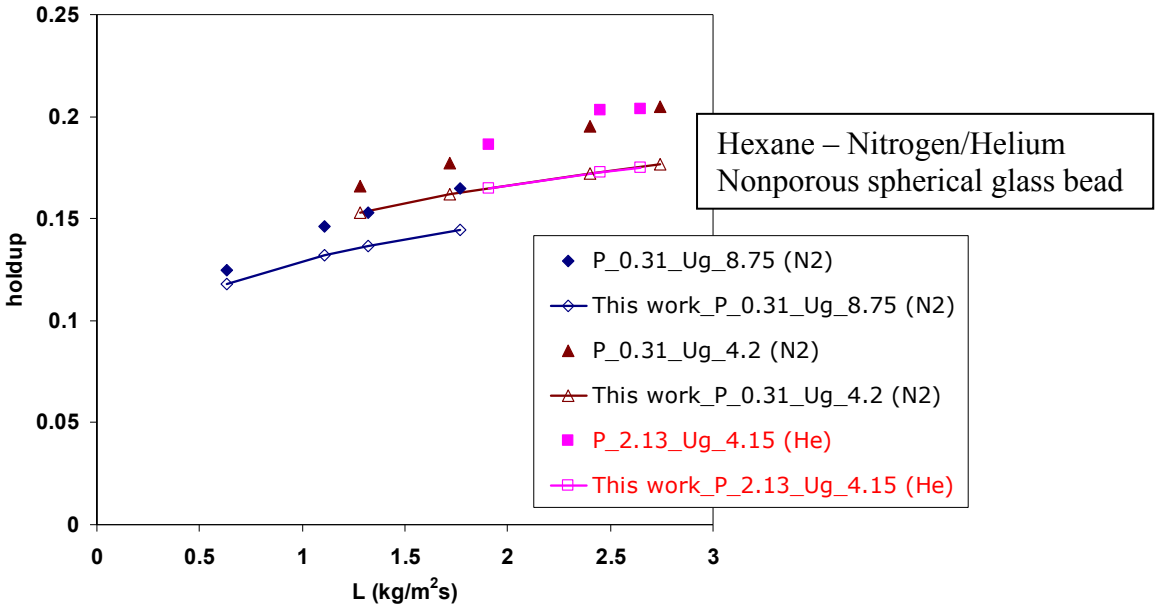

Fig 6: Comparative study of effect of gas density and flow rate on liquid holdup with Al-Dahhan and Dudukovic (1994)

It can be seen from these figures and that for a system with nitrogen as gas phase and with 0.31 MPa operating pressure exhibits almost same pressure drop and liquid holdup as the in the case for a system with helium as gas phase and with 2.13 MPa operating pressure. This interesting result is also evident from the numerical predictions of this work. The reason behind this phenomenon is that the density of helium at 2.13 MPa is equals to that of nitrogen at 0.31 MPa pressure. Therefore both the gas phases behave similar at those corresponding operating pressures for the same flow rate (in this case ≈ 4.2 cm s^{-1}).

It is established from the overall force balance on the gas phase that the pressure gradient is proportional to the gas–liquid interfacial drag (please refer to Equation 2). Besides the bed characteristics, this pressure gradient depends on the velocities of both phases and also the physicochemical properties of the flowing fluids (Saroha et al.,1998). In case of high pressure operation, with the increase of pressure, only gas density changes of all physicochemical properties of the flowing fluids. The body force depends on liquid density which is not substantially affected by pressure in the usual operating range of TBRs (less than 30MPa). Therefore, the effect of gas phase on high pressure hydrodynamics can be perceived in two ways: (a) effect of the superficial gas velocity and (b) gas density. For a given set of gas and liquid velocities, increased gas density leads to increased gas–liquid interaction and thus higher pressure drop. Both the cases have been validated here with sufficient degree of accuracy.

However, observing relatively higher deviation from the experimental data in cases of Figs 5 and 6, we have carried out some experimentations by changing Ergun constants to A=150 and B=1.5 (as suggested by Nemec and Levec (2005) for two phase flow) and adopting the static liquid correlation from Nemec and Levec (2005). These changes have minimized the relative discrepancies between the predicted results and experimental observations which are evident from Figs 7 and 8.

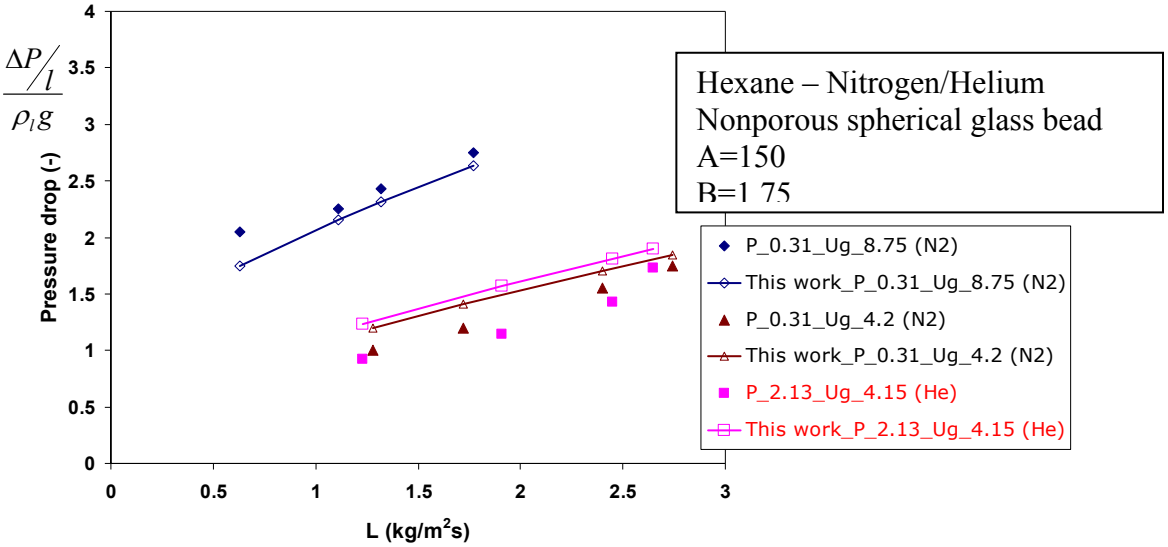

Fig 7: Comparative study of effect of gas density and flow rate on pressure drop with Al-Dahhan and Dudukovic (1994) for modified Ergun constants and static holdup correlation

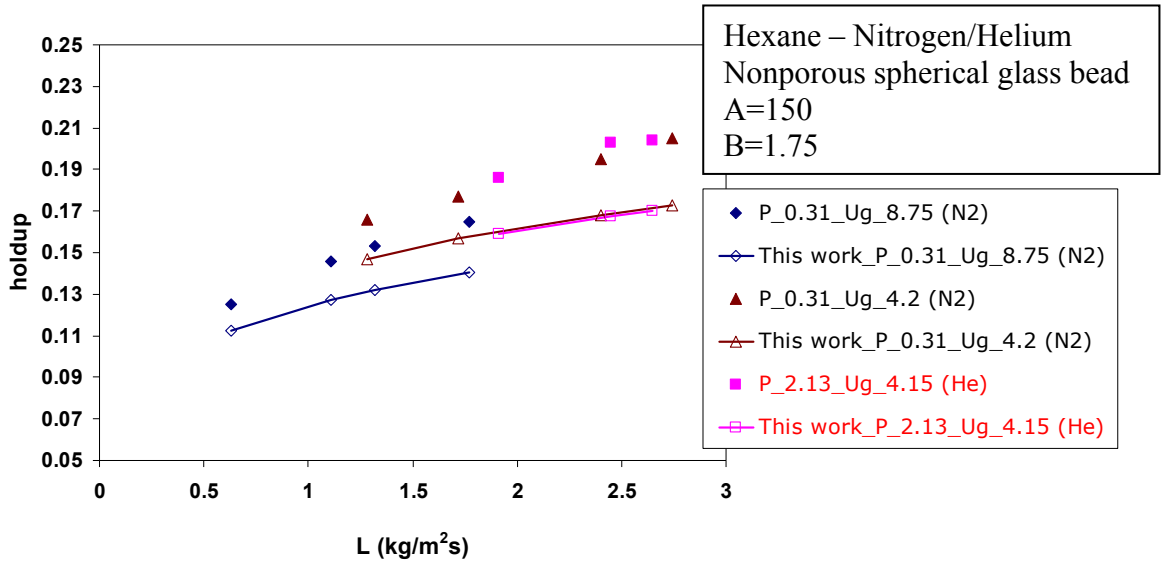

Fig 8: Comparative study of effect of gas density and flow rate on liquid holdup with Al-Dahhan and Dudukovic (1994) for modified Ergun constants and static holdup correlation

5. Summary and Conclusions

A less computationally intensive, yet first-principle based CFD model has been presented in this work using the porous media concept. While simulating for high pressure condition, we have applied the recently developed correlations by Nemec and Levec (2005) for relative permeabilities. The developed model is very much flexible unlike the traditional CFD approach, i.e. three-phase Eulerian simulations for different particle size effect incorporation without much complexity. It also seems to be a promising alternative to multi-fluid Euler-Euler drag models for trickle bed reactors. This CFD model can productively be implemented for high pressure operation (most of the commercial TBRs operate) which is cumbersome to account for three-phase Eulerian simulation. It has also been observed that use of proper Ergun's Constants is necessary for more accurate predictions to incorporate the catalyst particle shape effect in the developed CFD model. To quantify the overall accuracy of this model, we have also calculated Mean Average Relative Error (MARE; for details Al-Dahhan and Dudukovic, 1994) on the basis of 49 simulated data points for pressure drop and 45 simulated data points for liquid holdup from Al-Dahhan and Dudukovic (1994) which appear to be 23.46% and 15.08% for pressure drop and liquid holdup respectively. Therefore, it can be concluded that our model functions arguably well in predicting the experimental data in most cases. In future, this model can be tested with the results from a *real* trickle bed reactor for further investigations and robustness.

As high-pressure operation processes in TBRs are very frequently encountered in petroleum refining industry which is one of the most critical and biggest industry in the globe, any optimistic contribution towards its design and or modeling of different

parameters will not only lead to technology development thus resulting in substantial savings but will also help to maintain a cleaner and greener environment.

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Nomenclature

A	constant in the viscous term of the Ergun type equation
A_p	particle surface area, (m^2)
B	constant in the inertial term of the Ergun type equation
d_e	equivalent particle diameter, $\frac{6V_p}{A_p}$
Eo^*	modified Eötvos number, $\frac{\rho_l g d_p^2 \varepsilon^2}{\sigma_l (1-\varepsilon)^2}$
F_α	drag force on the α phase per unit volume, (kg/m^2s^2)
g	gravitational acceleration, (m/s^2)
Ga_α	Galileo number of the α phase, $\frac{\rho_\alpha^2 g d_e^3 \varepsilon^3}{\mu_\alpha^2 (1-\varepsilon)^3}$
k_α	relative permeability of α phase
l	length of the reactor, (m)
p	pressure, (Pa)
Re_α	Reynolds number of the α phase, $\frac{\rho_\alpha \varepsilon_\alpha u_\alpha d_e}{\mu_\alpha (1-\varepsilon)}$
s_α	saturation of α phase
u	superficial velocity, (m/s)
V_p	particle volume, (m^3)

Greek letters

δ_l	reduced saturation of liquid phase, $\frac{\varepsilon_l - \varepsilon_l^0}{\varepsilon - \varepsilon_l^0}$
ε_l^0	static liquid hold up
ε	bed voidage

ε_α	hold-up of α phase
μ	viscosity (Pa.s)
ρ_α	density of α th phase (kg/m^3)
σ	surface tension (N/m)

Subscripts

α	gas/liquid phase
g	gas phase
l	liquid phase

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