CFD Simulation of Multiphase Flow in Trickle Bed Reactor

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***Abstract*— The design and scale-up of trickle bed reactors(TBR) depend on key hydrodynamic variables such as reduced liquid saturation and pressure drop, which in turn depend upon parameters such as operating pressure, phase velocity and phase density. Most of the previous studies on quantifying reduced liquid saturation and pressure drop have been conducted under atmospheric pressure, whereas industrial operating pressures are around 20-30 MPa. Therefore, in the present study, using the bed dimensions and different operating conditions prescribed by Al-Dahhan and Dudukovic(1994) at high pressure, unsteady state behaviour of TBR has been investigated using commercial computational fluid dynamics (CFD) code Fluent based on the porous media concept developed by Atta et al. (2007). A comparative study, of three drag models (Sáez and Carbonell(1985), Nemec and Levec(2005), modified Nemec and Levec), at high pressure conditions is done. The predicted data for these drag models is compared to the experimental data from Al-Dahhan and Dudukovic (1994) and it was found that there are different zones in which different drag models gives accurate results. Also, the effect of operating parameters on hydrodynamics of TBR for these three drag models is studied at high pressure conditions. Later, from the overall zones, it was found that Sáez and Carbonell drag model suits best. Using this drag model, the results are then validated with Atta et al., at atmospheric pressure condition also. The present study concludes that the present model is valid for high pressure as well as low pressure conditions.**

***Keywords*— Trickle bed reactor, high pressure, porous media, hydrodynamics, pressure drop, CFD**

I. Introduction

A trickle-bed reactor (TBR) is a fixed bed of catalyst contacted by co-current downflow of gas and liquid. It is used widely in petroleum, petrochemical, and chemical industries. The design and scale-up of trickle bed reactors depend on key hydrodynamic variables such as liquid saturation, particle wetting and overall gas–liquid distribution. Experimental determination of these variables is difficult as the interactions between these parameters are poorly understood. Increasing computational power and recent development of Computational Fluid Dynamics (CFD) has made it possible to simulate multiphase flow in TBR [3,11,8]. The previous attempts for describing trickle bed hydrodynamics can be categorized mainly into two different classes of work. The classical approach is empirical wherein correlations are developed to 'fit' the experimental data [6,10]. Another approach is to describe hydrodynamics in phenomenological manner, i.e., assuming a simple picture of the microscale flow pattern, and then integrate that depiction to address the entire bed [9].

II. Previous Work

In recent years, many works devoted to the numerous aspects of behavior in fixed-bed reactors, such as hydrodynamics, chemical kinetics, mass and heat transfer have been published [1-12]. However, only in the last decades particular attention has been given to the behaviour of pressurized TBRs [7,10] whereas, the other authors [8,11] dealt with TBR research at atmospheric conditions. Various correlations and semi-theoretical models for prediction of two-phase pressure drop and liquid holdup at high-pressure operation have been summarized by Al-Dahhan *et al.* (1997). No correlation emerges as clearly superior to others, but those based on semi-theoretical and phenomenological models seem more reliable than strictly empirical correlations[1,3,9]. In trickle flow regime, the slit [9], and double slit [10] models, and the 1-D CFD model of Attou *et al.* (1999) are recommended for use to predict liquid holdup. Recently, Boyer *et al.* (2007) established a large experimental database at "Institut Francais du Petrole" to measure simultaneously pressure drop and liquid holdup in packed bed reactor operated in trickle flow regime for a large range of operating conditions. Nigam and Larachi (2005) and Saroha and Indraneel (2008) have reported that hydrodynamic hysteresis may occur at high pressure when the liquid is contaminated with impurities. The interstitial nature of liquid flow in trickle beds has become available due to the increased use of non-invasive sophisticated measurement techniques such as capacitance tomography imaging to capture the transient pattern of liquid flow in a trickle bed (Sederman and Gladden, 2005).

III. Present Work

As discussed above, most of the previous studies on quantifying reduced liquid saturation and pressure drop have been conducted under atmospheric pressure, whereas, the desired conditions of investigation are industrial operating pressures of 20-30 MPa. 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, the present work is lead by the fact that any optimistic contribution towards its design via 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. A comparative study of drag models (listed in Table I) is done to study the effect of various operating parameters such as reactor pressure, phase velocities, particle size on hydrodynamic parameters of trickle bed reactor. This present study is based on the 2D trickle bed geometry studied by various authors as listed in Table II. The bed dimensions are chosen correspondingly for validation of the present model. The bed consists of solid material of uniform diameter. Instead of specifying volume fraction of solids inside the bed (Gunjal et al., 2005), the solution domain is defined as porous. Numerical simulations were carried out for different geometries and flow conditions listed in Table II accounting uniform and constant porosity of the bed for simplicity of the computational model. Geometry is shown in Figure 1.

Table I Different Drag models used in this study

Drag Model I (Case I)	Relative permeabilities	Gas phase	$k_g = s_g^{4.80}$	Sàez and Carbonell (1985)
		Liquid phase	$k_l = \delta_l^{2.43}$	
	Static liquid holdup	$\varepsilon_l^0 = \frac{1}{\left(20+0.9Eo^*\right)}$ where	$Eo^* = \frac{\rho_l g d_p^2 \varepsilon^2}{\sigma_l (1-\varepsilon)^2}$	
	Ergun constants	A=180; B=1.8		
Drag Model II (Case II)	Relative permeabilities	Gas phase	$k_g = 0.4S_g^{3.6}(S_g \le 0.64)$ $k_g = S_g^{5.5}(S_g > 0.64)$	Nemec and Levec (2005)
		Liquid phase	$k_l = \delta_l^{2.9}(\delta_l \ge 0.3)$ $k_l = 0.40\delta_l^{2.1}(\delta_l < 0.3)$	
	Static liquid holdup	$\varepsilon_l^0 = 0.028\frac{1-\varepsilon}{\varepsilon}$		
	Ergun constants	A=150; B=1.75		
Drag Model III (Case III)	Relative permeabilities	Gas phase	$k_g = 0.4S_g^{3.6}(S_g \le 0.64)$ $k_g = S_g^{5.5}(S_g > 0.64)$	Nemec and Levec (2005)
		Liquid phase	$k_l = \delta_l^{2.9}(\delta_l \ge 0.3)$ $k_l = 0.40\delta_l^{2.1}(\delta_l < 0.3)$	
	Static liquid holdup	$\varepsilon_l^0 = \frac{1}{\left(20+0.9Eo^*\right)}$ where	$Eo^* = \frac{\rho_l g d_p^2 \varepsilon^2}{\sigma_l (1-\varepsilon)^2}$	Sàez and Carbonell (1985)
	Ergun constants	A=180; B=1.8		

Table II Details of operation conditions used for simulations at high pressure

Source	Operating Pressure (MPa)	System		Packing		
				Catalyst	Size[mm]	ε
Al-Dahhan and Dudukovic (1994)	0.3-5.0	Water, Hexane	Nitrogen, Helium	0.5% Pd/alumina	$(d_p)_{eq}$=1.99	0.355
				Silica shell(sphere)	d_p = 1.52	0.412
				Glass beads(sphere)	d_p = 1.14	0.392

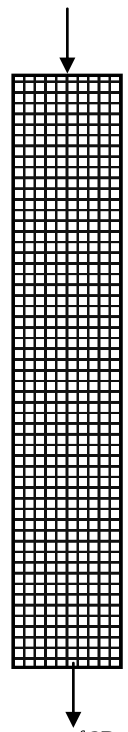


Fig. 1 Geometry of 2D trickle bed

IV. MODELING

The model presented here to describe the multiphase flow is based on the Eulerian framework, which consists of continuity and momentum equations of each fluid phase with appropriate drag models (Table 1).

Continuity equation:

$$\frac{\partial\left(\alpha_g \rho_g\right)}{\partial t}+\nabla\left(\alpha_g \rho_g \vec{\upsilon}_g\right)=0 \quad (1)$$

Momentum balance equation:

$$\frac{\partial\left(\alpha_g \rho_g \vec{\upsilon}_g\right)}{\partial t}+\nabla\left(\alpha_g \rho_g \vec{\upsilon}_g \vec{\upsilon}_g\right)=-\alpha_g \nabla P+\nabla\left(\bar{\tau}_g\right)+\alpha_g \rho_g \vec{g}+K_{gs}\left(\vec{\upsilon}_g-\vec{\upsilon}_s\right) \quad (2)$$

In order to solve these model equations the following assumptions are taken. 1. There is no inter-phase mass transfer. 2. Both the flowing fluids are incompressible. 3. The porous medium is taken to be isotropic, i.e., permeabilities are independent of direction. 4. The porosity is uniform and constant. 5. Trickle flow regime is the operating flow regime, i.e., gas-liquid interaction is low so capillary pressure forces can be neglected. This means that same pressure is assumed for both phases at any point in time and space and 6. The contribution of the turbulent stress terms to overall momentum balance equation (Eqn. 2) is not significant. This assumption has also been used by other authors (e.g. Jiang et al., 2002[a]).

V. BOUNDARY CONDITIONS AND NUMERICAL SIMULATION

Several numerical simulations were carried out and the simulated results were validated against the published experimental data[1] for pressure drop and reduced liquid saturation from an independent source (Al-Dahhan and Dudukovic, 1994). The details of the operating conditions for the experimental datasets (adopted from Al-Dahhan and Dudukovic, 1994) are shown in Table II. The bed dimensions were chosen according to the packed bed characteristics prescribed by Al-Dahhan and Dudukovic (1994) and Larachi et al. (1991).

Table III Simulation parameters

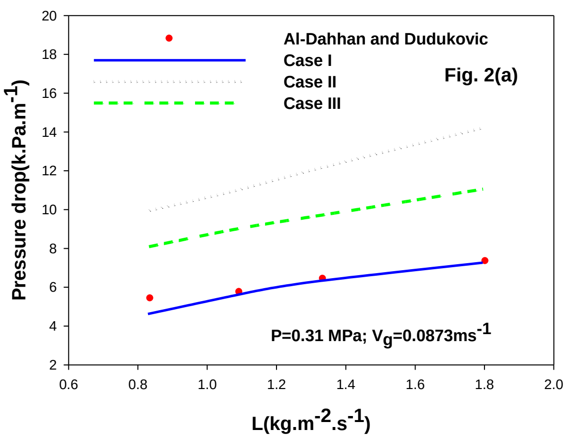
Under Relaxation Factors	
Pressure	0.3
Momentum	0.7
Volume Fraction	0.2
Body forces	1
Density	1
Model parameters	
Solver	Unsteady, 2nd order Implicit
Multiphase	Eulerian-Eulerian
Discretization scheme	First order Upwind
Pressure-velocity coupling	Phase coupled SIMPLE
Time step	0.005 s
Iterations Per Time step	20
Convergence criterion	10^{-5}

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. The reference pressure equal to atmospheric pressure was fixed at the outlet. Unsteady state simulations were carried out with the time step of 0.005 s. Some preliminary numerical tests were carried out to identify the required number of computational cells to obtain grid independent results. Further, it was also ensured through preliminary numerical tests 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. The simulation parameters are summarized in Table III.

VI. Results And Discussion

A comparative study of the drag models listed in Table I at high pressure (3.91 MPa) and the effect of various output parameters with variation in input parameters is discussed in this section.

A. *Effect of operating pressure on hydrodynamics*



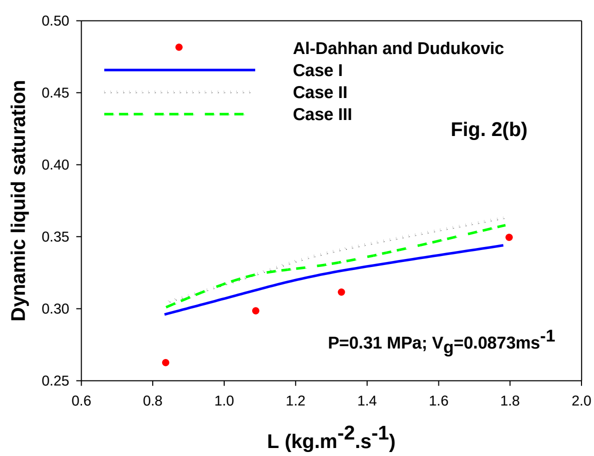
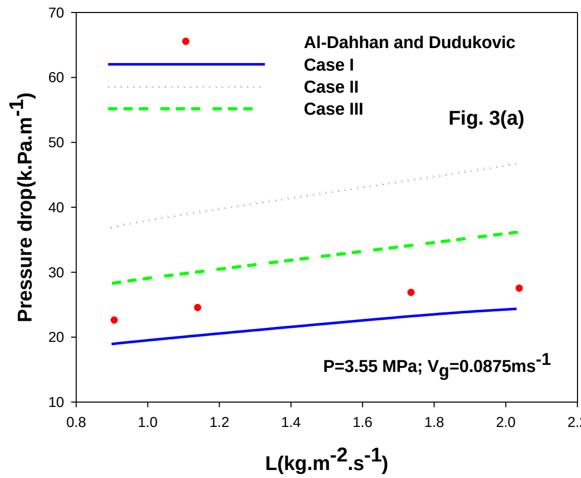


Fig 2 Comparative study of (a) pressure drop and (b) reduced liquid saturation with literature data[1]; system: hexane–nitrogen; 0.5% Pd on alumina.

Figs. 2(a) & 3(a) represent the comparative studies of effect of reactor pressure for two different operating pressures (0.31 MPa and 3.55 MPa) on pressure drop as a function of Liquid mass flux (L) with experimental data of Al-Dahhan and Dudukovic (1994) at a constant



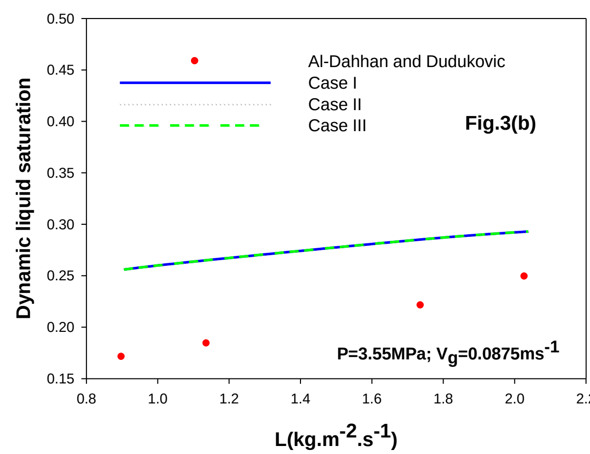


Fig. 3 Comparative study of (a) pressure drop and (b) reduced liquid saturation with literature data[1]; system: hexane–nitrogen; 0.5% Pd on alumina; P=3.55MPa, V_g=0.0875 ms^{-1}

gas velocity of 8.75 $cm.s^{-1}$ for a system of hexane-nitrogen with 0.5% Pd/alumina porous extrudate catalyst. Similarly, Figs. 2(b) and 3(b) show the effect of reactor pressure on reduced liquid saturation as a function of Liquid mass flux (L). Comparing these figures, it is observed that for a particular gas velocity (0.0875 ms^{-1}), with the increase in operating pressure, pressure drop increases as can be seen from Figs. 2(a) & 3(a) whereas, corresponding reduced liquid saturation decreases as is observed from Figs. 2(b) & 3(b). Drag Model I shows better agreement with the experimental pressure drop with an error between 3% to 15% for Figs. 2(a) & 3(a), but it overpredicts the reduced liquid saturation (error between -19% to -4.4% for Figs. 2(b) & 3(b)). The other drag models (Drag Model *II* and *III*) are not in good agreement even for pressure drop (error between -21% to -7.9% for Drag Model III and -26% to -7.1% for Drag Model II). Interestingly Drag Model I, Drag Model II and Drag Model III predict almost same values for reduced liquid saturation.

The reason behind this observation is that at higher pressure, there is a larger amount of gas-liquid interfacial interaction, which results in higher pressure drop values as it is established from the overall force balance on the gas phase that the pressure gradient is proportional to the gas–liquid interfacial drag (Eq. (2)). It is noteworthy that the range of liquid and gas velocities used here as operating conditions (Table II) are 65-75% of the velocity at flooding condition.

B. Effect of liquid phase and gas phase materials on hydrodynamics

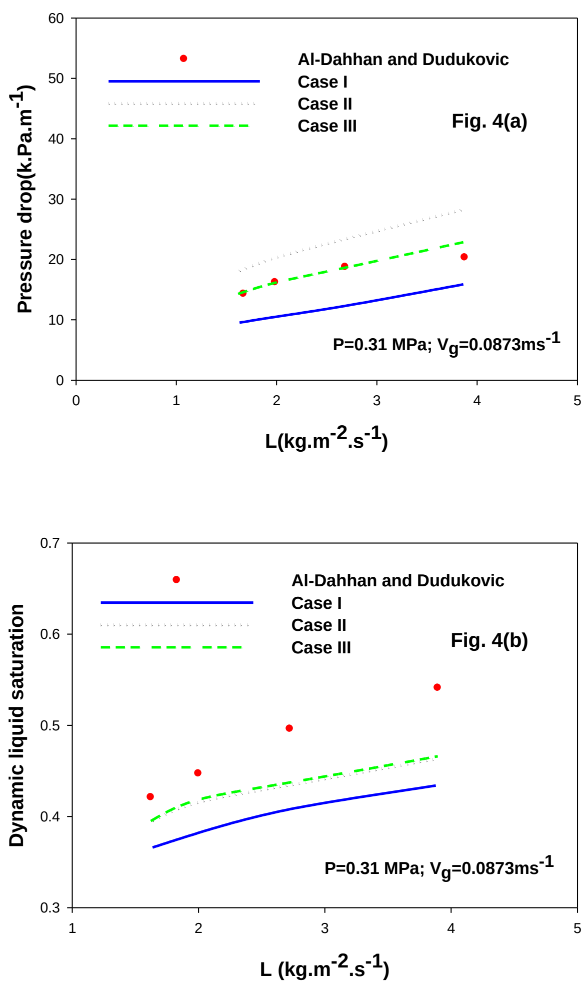


Fig. 4 Comparative study of (a) pressure drop and (b) reduced liquid saturation with literature data; system: water–nitrogen; 0.5% Pd on alumina.

Comparing the Figs. 2 & 4, it can be observed that for a particular velocity (0.0875 ms^{-1}) and operating pressure of 0.31 MPa, at a Liquid mass flux of 1.8 kg/m^2s, hexane-nitrogen system shows a pressure drop of 6 $k.Pa.m^{-1}$ and reduced liquid saturation of 0.34 whereas, water-nitrogen system shows a pressure drop of 12 $k.Pa.m^{-1}$ and reduced liquid saturation of 0.42. The reason behind this observation is the increase in liquid density which substantially affects the hydrodynamics. For this case, Drag Model III shows better agreement with the experimental data with an error of -12.6% to -3.2%.

C. Effect of gas velocity on hydrodynamics

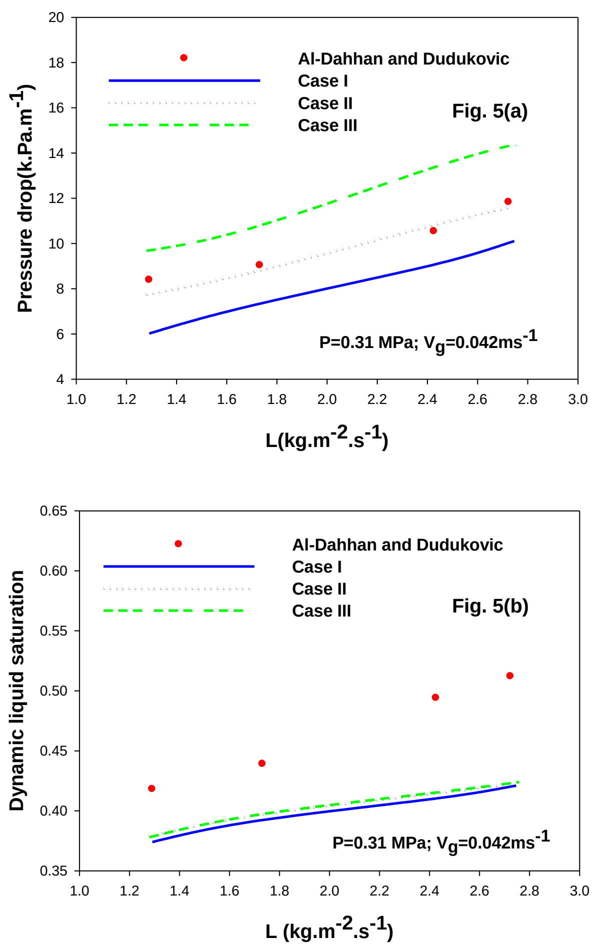


Fig. 5 Comparative study of (a) pressure drop and (b) reduced liquid saturation with literature data; system: hexane–nitrogen; glass bead.

Comparing the Figs. 5 & 6, it can be observed that for a particular operating pressure of 0.31 MPa, at a Liquid mass flux of 1.8 kg/m^2s, Fig 5 shows a pressure drop of 8 $k.Pa.m^{-1}$ and reduced liquid saturation of 0. 4 whereas, Fig 6 shows a pressure drop of 15 $k.Pa.m^{-1}$ and reduced liquid saturation of 0.35.

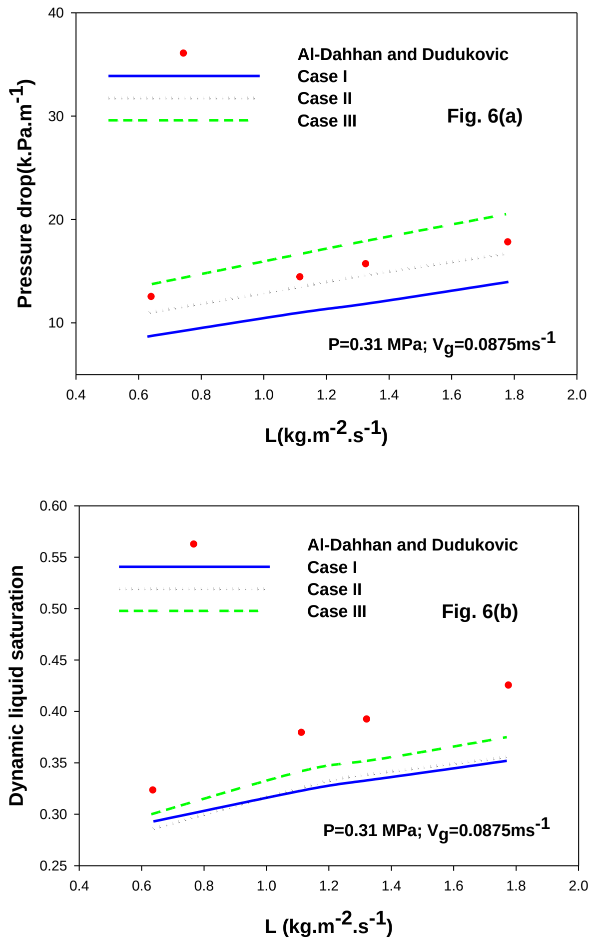


Fig. 6 Comparative study of (a) pressure drop and (b) reduced liquid saturation with literature data; system: hexane–nitrogen; glass bead.

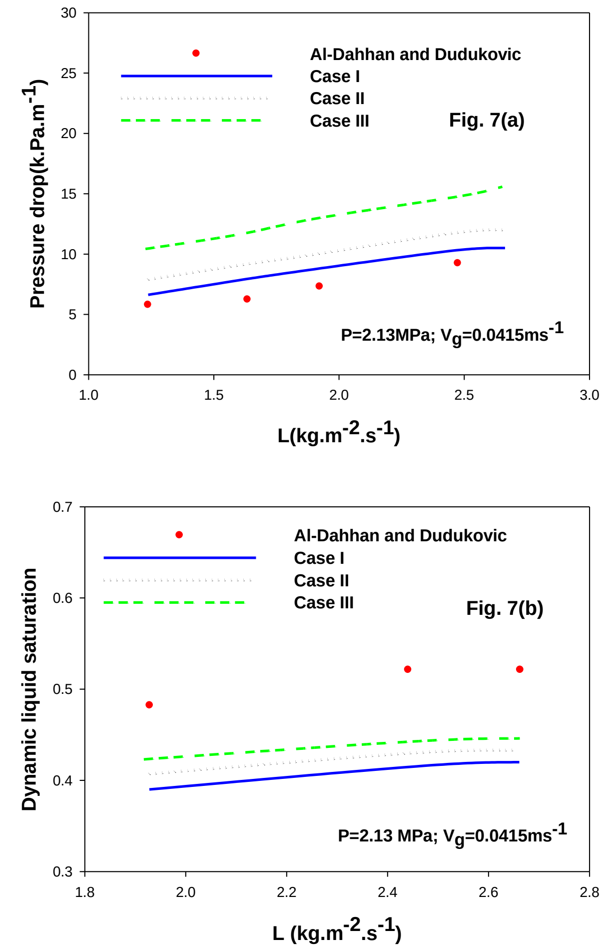


Fig. 7 Comparative study of (a) pressure drop and (b) reduced liquid saturation with literature data; system: helium–nitrogen; glass bead.

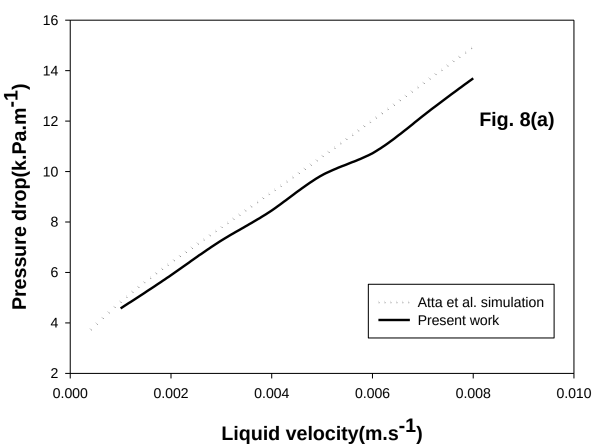
The reason behind this observation is that at higher velocities, due to transition in flow regime, there is a larger amount of gas-liquid interfacial interaction, which results in higher pressure drop values. Figs 5 & 6 shows that the better fit is achieved when the Drag Model *II* (error between -1% to +6%) is applied in the model. In line with the experimental results (Table II), it also shows that for a constant operating pressure (0.31MPa), increasing gas velocities result in higher pressure drop and lower reduced liquid saturation. The reason for this observation is discussed above.

D. Effect of gas density on hydrodynamics

It can be seen from the Figs. 5(a) & 7(a) that a system with nitrogen as gas phase and with 0.31MPa operating pressure exhibits almost same pressure drop as in the case for a system with helium as gas phase and with 2.13MPa operating pressure when liquid phase is varied as well. Similar observation is made from Figs. 5(b) & 7(b) for reduced liquid saturation. The reason behind this observation is that the density of helium at 2.13MPa ($\sim$3.44 kgm^{-3}) is almost equal to that of nitrogen at 0.31MPa pressure ($\sim$3.51 kgm^{-3}). Therefore both the gas phases behave similarly at these corresponding operating pressures for the same velocity (in this case $\approx$4.2cms^{-1}). From the Figs. 7(a) & 7(b), it is evident that Drag Model I exhibits better fit (error between -3% to +15%) amongst all in these drag models investigated.

From the above observations,, it can be concluded that the present simplified CFD model to forecast hydrodynamics of high-pressure TBR functions performs well in predicting a wide range of operating condition (as given in Table II) including different particle shape and size of the packing. To quantify the overall accuracy of this model, Mean Average Relative Error (MARE) is also calculated on the basis of simulated data points for pressure drop and reduced liquid saturation from the predicted data, and the best selection seems to be Drag Model *I* in which percentage errors appear to be 19.69% and 18.76% for pressure drop and reduced liquid saturation respectively. Using this Drag Model I, validation of present results is carried out with that of Atta et al., (2007).

E. Validation of present simulation results with that of Atta et al.



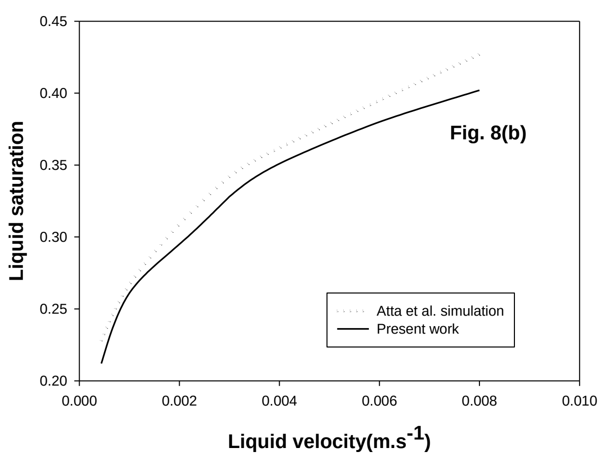


Fig. 8 (a), (b) Comparative study with Atta et al. (V_g=0.22 ms^{-1})

Figs. 8(a) & 8(b) show the comparison of simulated results of present study with that obtained by Atta et al. (2007) through simulation for observed pressure drop and reduced liquid saturation respectively. Comparing these figures, it can be observed that pressure drop increases with liquid velocity whereas, reduced liquid saturation decreases. The predicted values are very closely matching to simulation results of Atta et al. at low liquid velocity.

The reason for the second observation is that at high liquid velocity, the porosity distribution changes in the bed and there is no readily available method for implementing porosity profile in the present model and therefore uniform porosity throughout the bed is assumed. However, Atta et al. in their simulation have assumed an ad hoc porosity profile proposed by De Klerk (2003). The maximum error between the present work and that of Atta et al., is found to be +15%.

VII. CONCLUSIONS

In the present work, unsteady state behaviour of trickle bed reactor has been investigated. A two-phase Eulerian CFD model based on porous media concept to simulate gas–liquid flow through trickle bed is developed. The following conclusions can be drawn from the results:
The pressure gradient along the length of TBR depends on the velocities of both phases and also on the physicochemical properties of the flowing fluids specially density. With the increase in velocities of both phases, the pressure drop per unit length increases.

1. For a particular gas velocity, with the increase in operating pressure, pressure drop per unit length increases whereas corresponding reduced liquid saturation decreases.
2. In case of high-pressure operation, with the increase of pressure, only gas density changes significantly out of all physicochemical properties of the flowing fluids whereas, liquid density is not affected much as the pressure in the usual operating ranges of TBRs are less than 30MPa.
3. The effect of gas phase on high-pressure hydrodynamics can be perceived in two ways: effect of (a) superficial gas velocity and (b) gas density. For a given set of gas and liquid velocities, increased gas density leads to increased Reynolds number and thus results in a transition of flow. For a given system, with the increase in gas velocity, the pressure drop increases.
4. While predicting the hydrodynamics of high-pressure TBRs, it is found that when the gas phase saturation exceeds over 0.64 and reduced liquid saturation is below 0.25, Sáez and Carbonell(1985) drag model in general, perhaps appears to be better (error between -3% to +15%) amongst the drag models tested, For a particular set of operating condition, if the reduced liquid saturation varies between 0.25–0.38 or gas saturation is near the value of 0.64, Nemec and Levec (2005) drag model appears to be better (with an error between -1% to +6%).

5. The transition from rivulet liquid flow into liquid films was observed to initiate at liquid superficial velocities between 1.8 and 2 mm/s. It is observed that this transition phenomenon has affected the hydrodynamics of TBR at high velocities significantly.

NOMENCLATURE

A	Constant in the viscous term of the Ergun type equation
B	Constant in the inertial term of the Ergun type equation
d_e	Equivalent particle diameter, $6V_p/A_p$ (m)
d_p	Particle diameter (m)
Eo^*	Modified Eotvos number, $\rho_l g d_p^2 \varepsilon^2/(\sigma_l(1-\varepsilon)^2)$
g	Gravitational acceleration (m/s^2)
Ga_α	Galileo number of the α phase, $\rho_\alpha^2 g d_e^3 \varepsilon^3/(\mu_\alpha^2(1-\varepsilon)^3)$
k_α	Relative permeability of α phase
K_{gs}	Gas/solid momentum exchange (kg/m^{3}s)
L	Liquid mass flux (kg/m^{2}s)
P	Pressure (Pa)
Re_α	Reynolds number of the α phase, $\rho_\alpha \varepsilon_\alpha \mu_\alpha d_e/(\mu_\alpha(1-\varepsilon))$
S_α	Saturation of α phase, $\varepsilon_\alpha/\varepsilon$
$\overline{u}$	Fluid phase velocity (m/s)
V_g	Gas velocity (m/s)
	Greek Letters
δ_l	Reduced saturation of liquid phase, $(\varepsilon_l - \varepsilon_l^0)/(\varepsilon - \varepsilon_l^0)$
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)
$\vec{\upsilon}$	Instantaneous velocity vector (m/s)
$\overline{\tau}$	Phase stress-strain tensor (Pa)
	Subscripts
α	Gas/Liquid phase
g	Gas Phase
l	Liquid phase
TBR	Trickle Bed Reactor

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