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SIMULATE THE FLOW IN MONOLITH REACTORS USING CFD

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The objective of this research work is to simulate the flow in monolith reactors using computational fluid dynamics for both single channel and monolith bed. It proposes a co-current down flow monolith reactor system to investigate the flow distribution characteristics in a 2 inch monolith reactor equipped with selected types of distributors and operated in the Taylor flow regime. The effects of liquid distributor type and the liquid and gas velocities within the desired flow regime of Taylor flow on the flow distribution have also been investigated and provided. The existing single tube model for the Taylor flow regime has modified by integrating the hydrodynamics and kinetics to predict the performance of monolith reactor. The analysis on both single channel and monolith bed has been done and compared with the experimental data provided.

Keywords: Monolith reactor, CFD, Taylor flow, Single channel monolith, Hydrodynamics of monolith bed

1. Introduction

Monolith loop reactors are gaining considerable attention from academia and industry alike for carrying out solid catalyzed gas–liquid reactions. Monolith loop reactors are being applied in laboratory studies and in commercial practice for carrying out reactions such as hydrogenations, hydrodesulphurization, oxidations and Fischer-Tropschsynthesis. Monolith reactors offer many potential advantages over trickle beds, slurry bubble columns and airlifts that include low pressure drop, high mass transfer rates, and ease of scale up. Provided the gas and liquid phases are uniformly distributed over the various channels of the monolith, commercial reactor of large dimensions can, in principle, be scaled up from information on the hydrodynamics, mass transfer, and mixing within a single channel that has dimensions typically in the 1–3 mm range. Inside each capillary, we usually have Taylor flow of gas bubbles. In the development and design of monolith loop reactors for fast reactions, the mass transfer from the Taylor gas bubbles to the surrounding liquid phase becomes an important limiting factor, Van Baten & Krishna [3].

Monolith reactor is a tubular reactor stacked with catalyst coated monoliths instead of random packings. In a typical monolith parallel channels are separated by walls made of cordierite (magnesium aluminosilicate) or other ceramic materials. Monoliths can carry active catalysts in two ways: the surface can have a wash coat of the active catalyst, or the structure can be impregnated with active catalyst.

Monoliths are industrially produced by extrusion of a paste containing catalyst particles or by extrusion of a support on which the catalyst can be coated (wash coating). In monoliths the channel cross sections are usually rectangular but circles, triangles, hexagons or more complex geometries also exist. To increase the surface area internal fins can also be provided. These fins have a stabilizing effect on the gas liquid flow and allow operation in counter current mode without flooding, Heibel et al. [1] and Lebens et al. [5-8].

Monolith reactors can be operated in two distinct flow regimes, Taylor flow regime (or slug flow regime) and annular flow regime. Taylor flow regime is characterized by the movement of the train of alternate gas bubbles and liquid slugs through the monolith capillary channels. On the other hand, in annular flow regime the liquid falls on the sides of the capillary walls and the gas flow through the core. But the preferred flow regime is Taylor or slug flow regime because monolith exhibits superior mass transfer characteristics

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when operated in this flow regime and also in this research Taylor flow regime is used, Roy [11].

Van Baten & Krishna [4] used computational fluid dynamics to investigate the mass transfer from the liquid phase to the channel wall for Taylor flow of bubbles rising in circular capillaries. The separate influences of the Taylor bubble rise velocity, unit cell length, gas holdup, and liquid diffusivity on mass transfer were investigated for capillaries of 1.5 mm, 2 mm and 3 mm diameter. A correlation was proposed for estimation of the wall mass transfer coefficient and this correlation had been tested against published experimental data.

Van Baten & Krishna [4] used computational fluid dynamics (CFD) to investigate mass transfer from Taylor bubbles to the liquid phase in circular capillaries. The liquid phase volumetric mass transfer coefficient k_{La} was determined from CFD simulations of Taylor bubbles in up flow, using periodic boundary conditions. The separate influences of the bubble rise velocity, unit cell length, film thickness, film length, and liquid diffusivity on k_{La} were investigated for capillaries of 1.5 mm, 2 mm and 3 mm diameter.

Liu et al. [2] introduced a novel structured metallic catalyst that improved mass transfer performance of a monolith reactor for highly exothermic gas–solid reactions. The monolith channels were designed to have metallic substrates that consist of two layers with one of the layers being the metallic support and another layer being a foam metal annular that was tightly deposited onto the support surface by some means. Parametrical studies based on a 2D monolith reactor model showed that the present design yielded an enhanced mass transfer between the bulk fluid and the catalyst layer due to a decrease in external film resistance, and an enhanced mass transfer within the solid phase mainly due to the viscous flow effect within the porous catalyst layer.

Natividad et al. [10] performed selectivity and kinetic studies of the Pd catalysed hydrogenation of 2-butyne-1,4-diol in a single capillary channel, and monoliths consisting of 1256 capillaries and 5026 capillaries in pressure range 100–300 kPa and temperature range 298–328 K using a 30% v/v 2-propanol/water solvent. All reactors were operated in down flow mode such that the reaction fluid was in Taylor flow.

Al-Dahhan [9] designed and developed an Industrial Tomography Scanner (ITS) to study and quantify the phase distribution in a two-phase flow pilot scale monolith reactor that was 24 in. (0.60 m) in diameter and 192 in. (4.9 m) in height. The monolith reactor was operated co-current up-flow in the Taylor flow regime with water as the liquid phase and air as the gas phase.

2. CFD Models

In first set of simulations, the work done by Krishna and Van Baten [3] using CFD Software CFX was reproduced by using CFD Software FLUENT for single channel analysis. In this analysis the bubble was held stationary and the wall moves down with velocity equal to bubble rise velocity. The dimensions of capillary were taken as same as that by Krishna and Van Baten [3].

In second set of simulations, the case of single capillary two phase system where the concurrent flow of water and air through the single capillary of monolith bed reactor was studied. In this case exact dimensions of Roy [11] were used.

In third set of simulations, the analysis of entire monolith reactor was carried out using porous media model for monolith bed of a monolith reactor in FLUENT.

2.1. Models information

In single channel analysis 2ddp axi-symmetric model was used. Dimensions used were diameter of capillary d_c = 3 mm, unit cell length = 40 mm, film thickness = 0.048 mm, length of liquid film = 5.31 mm, diameter of bubble = 2.904 mm, length of bubble = 8.214 mm, length of liquid slug = 31.786 mm. Solver used was segregated, steady, laminar. Boundary Conditions were periodic boundary condition at inlet and outlet, $U_{Top} = U_{Bottom}$, $P_{Top} = P_{Bottom}$. Bubble rise velocities (V_b) of 0.55 m/s , 0.45 m/s , 0.3 m/s , 0.2 m/s and 0.15 m/s were used. At the outer wall boundary condition were $U_X = V_{Wall} = - V_b$, $U_Y = 0$. The bubble surface is specified as free slip.

In single capillary two phase system 2ddp axi-symmetric model was used. Dimensions used were diameter of capillary = 1.253 mm, height of capillary = 15 cm, diameter of bubble = 1.19035 mm, number of channels = 1100 and open frontal area OFA = 75%. Solver used was segregated, unsteady and laminar. Approach used was Eulerian. Boundary conditions were velocity inlet for both gas and liquid inlets, pressure outlet at outlet. Liquid superficial velocities used for various runs are 0.025 m/s, 0.1m/s, 0.2 m/s. Gas superficial velocities used were 0.1 m/s and 0.2 m/s.

In monolith bed analysis using porous media model 2ddp axi-symmetric model was used. Dimensions used were diameter of monolith column = 5 cm, diameter of monolith bed = 4.8 cm, height of monolith bed = 15 cm, height of monolith reactor = 50 cm, diameter of bubble = 1 mm, hydraulic diameter = 1.1 mm, number of channels = 1100 and open frontal area OFA = 75%. Solver used was segregated, unsteady and k-ε model of turbulence was introduced for incorporating the turbulence effects. Approach used was Eulerian. Boundary conditions were velocity inlet for both gas and liquid inlets through nozzle with the help of user defined functions, pressure outlet at outlet. Liquid superficial velocities used for various runs are 0.1 m/s, 0.2 m/s. Gas superficial velocities used were 0.1 m/s, 0.2 m/s, 0.5 m/s. Porous media boundary condition was introduced for capillaries in monolith bed. Figure 1 shows grid and boundary conditions for all of the above cases.

3. Results of CFD Simulations

Figure 2 shows the velocity profile of water in single capillary monolith, which indicates the flow pattern around the bubble and recirculation in some regions. Figure 3(a) and figure 3(b) represents contours and velocity vectors of single channel of monolith with film portion being enlarged. A graph of bubble rise velocity and sum of liquid and gas superficial velocities representing the comparison of CFD results and results of Van Baten & Krishna [3] is shown in figure 4. Figure 5 represents the comparison of CFD results and results of Van Baten & Krishna [3] for film surface velocity and bubble rise velocity. The CFD results deviate a bit from the results of Van Baten & Krishna [3] but the trends are almost similar. One reason could be the assumption of 2d domain instead of 3d as phenomena in monolith is purely 3d, 2nd reason could be the assumption of steady state, 3rd reason is the assumption of no shear on wall. In figure 4, the value of total superficial velocity (U_g + U_l) is slightly smaller than the value of the bubble rise velocity, V_b and this is due to the backflow of liquid through the film. The difference between two results indicates that back flow is larger in case of FLUENT simulated results.

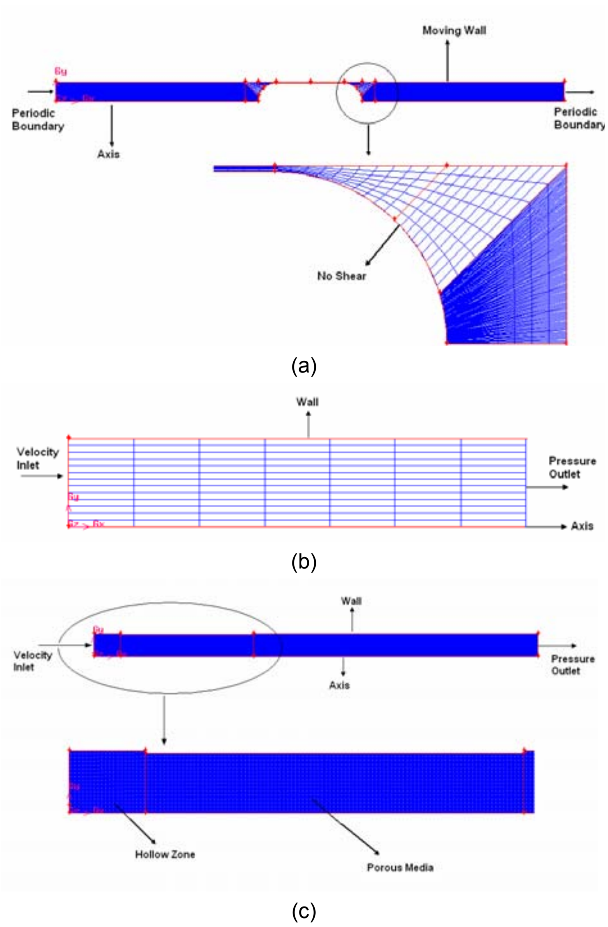


Figure 1 (a) 2ddp-axi-symmetric domain showing boundary conditions and grid of single capillary of monolith reactor (b) 2ddp-axisymmetric domain showing boundary conditions and grid of single capillary of monolith reactor (c) 2ddp-axisymmetric domain of monolith reactor showing boundary conditions and grid.

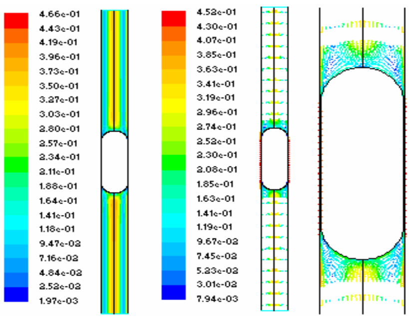
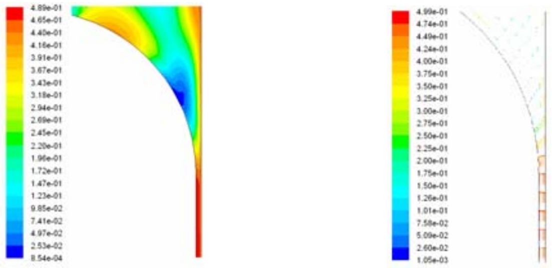


Figure 2. Contours of velocity magnitude of monolith for single phase in single capillary.



(a) (b)

Figure 3 (a) Contour of velocity magnitude of monolith for single phase in single capillary (film portion being enlarged). (b) Velocity vector of monolith for single phase in single capillary (film portion being enlarged).

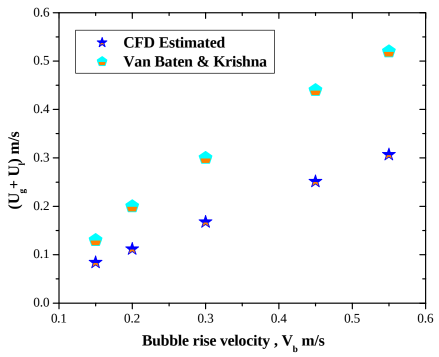


Figure 4. Graph of bubble rise velocity vs. total superficial velocity (U_g+U_l) m/s.

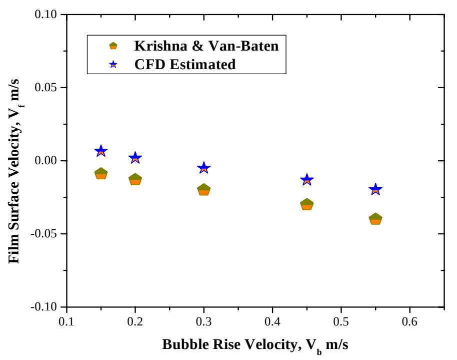


Figure 5. Graph of bubble rise velocity and film's surface velocity.

Figure 6 represents the contours of gas hold up of air in single capillary of monolith reactor, the contours of velocity vectors are also shown alongwith these as well. These contours show uniform phase distribution in a single capillary. Figure 7 represents the graphs of radial position against liquid saturation in a single capillary for various liquid and gaseous superficial velocities. It was then compared with the experimental data of Roy [11]. From this plot it is clear that significant uniformity in phase distribution has been achieved across radial position in case of single capillary. In literature it is mentioned that if 100% uniformity is achieved then single channel results can be used or scaled up for entire monolith reactor.

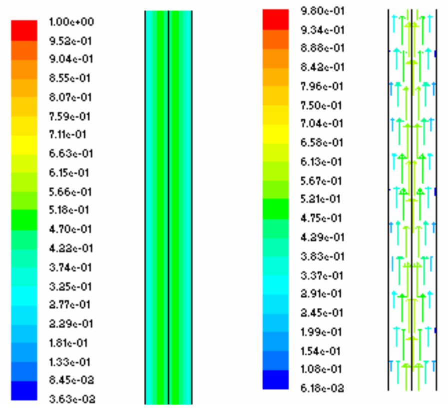


Figure 6: Contours of gas holdup of air in single capillary of monolith.

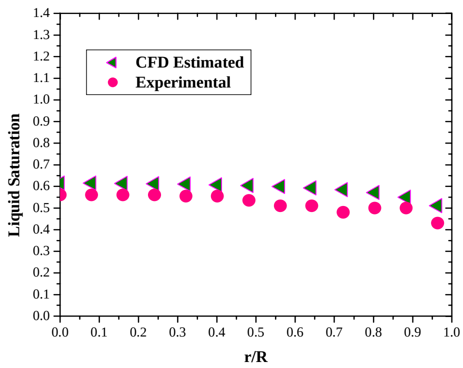


Figure 7: Graph of radial position vs. liquid saturation for Ul = 0.1 m/s and Ug = 0.2 m/s.

Figure 8 represents the contours of gas holdup for porous media of monolith bed alongwith the vector diagram of velocity magnitude. These contours indicate that due to the recirculations caused in the region before monolith bed air and water are not properly distributed throughout the bed. As a result of which almost all of the water passes through the middle portion of the monolith bed and the air passes through the sides of the monolith bed. Figure 9 represents graph of radial position against liquid hold up for various liquid and gaseous superficial velocities for the entire monolith bed of the monolith reactor. The monolith bed is modeled by applying porous media model. A significant flow mal-distribution is observed when graphs are plotted and compared with the experimental data of Roy [11]. The reason for this flow mal-distribution was high recirculations which were observed in the region of the monolith reactor which was present before the start of monolith bed. As a result of these recirculations uniform phase distribution was not achieved in monolith bed despite of using user defined functions for velocity inlet boundary condition.

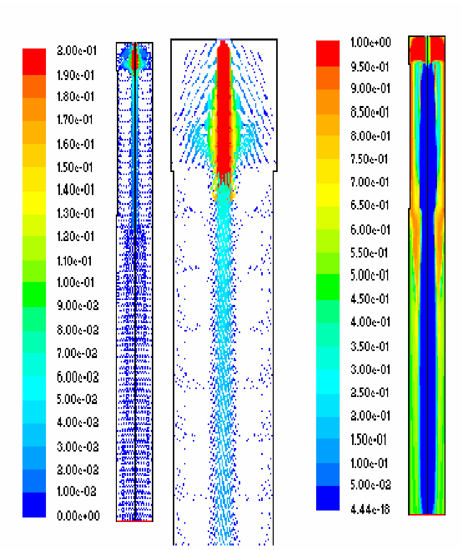


Figure 8. Contours of gas holdup for porous media of monolith bed.

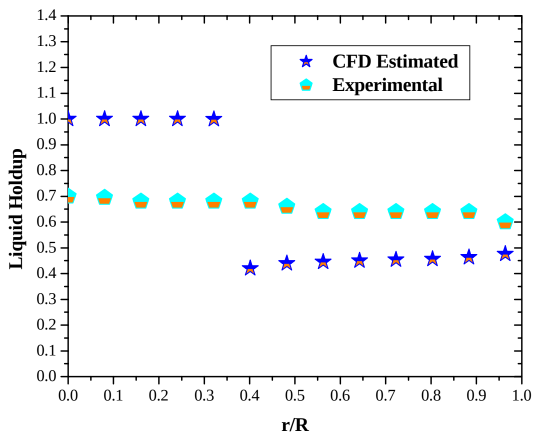


Figure 9. Graph of radial position vs. liquid holdup for U_l = 0.1 m/s and U_g = 0.1 m/s

4. Conclusions

The conclusions for various sections of this research can be summarized as follows for the three different cases being discussed. The results of single channel analysis were found to be in close agreement with results of Krishna & Van Baten [3]. The results of single capillary analysis for two phase flow through capillary showed that flow through the single capillary was laminar and uniformly distributed. The results of simulations carried out for whole monolith reactor using porous media model didn't match well with the experimental results because of recirculations in the region before monolith bed. It was found that liquid distributor type and its position above the monolith bed effects uniformity of phase distribution through monolith bed very significantly.

Notations

CFD	Computational fluid dynamics
d_c	Diameter of capillary
ITS	Industrial tomography scanner
k	Kinetic energy
k_{La}	Liquid phase volumetric mass transfer coefficient
OFA	Open frontal area
P_{Top}	Pressure at top
P_{Bottom}	Pressure at bottom
Pd	Palladium
U_g,	Superficial velocity of gas
U_l	Superficial velocity of liquid
U_X	Velocity of water in X-direction
U_Y	Velocity of water in Y-direction
U_{Top}	Velocity of water at top
U_{Bottom}	Velocity of water at bottom
V_{wall}	Velocity of wall
ε	Dissipation rate
2d	Two dimensional
2ddp	Two dimensional double precision

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