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Hydrodynamics characteristics of HDS trickle bed test reactor

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Abstract

Laboratory-scale trickle bed reactors are often used in testing of catalyst life time and activity. One of the important processes is represented by hydrodesulphurization (HDS) of motor fuels and petrochemical fractions. The study of stability of catalysts activity can be influenced both reactions kinetics, hydrodynamics and mass transfer phenomena. In case of ideal behaviour, the liquid could report piston flow. Non-ideal liquid flow can lead to incomplete catalyst particle wetting, which results in poor utilization of the catalytic surface of the particles. It may also expected to occur stagnant zones and imperfect heat transfer and overheating of the catalytic particles. For limitation of this negative phenomena should be diluted catalytic bed by small inert particle provide more contact area. The aim of this study was to determine degree of the effect of hydrodynamic conditions in model trickle bed reactors by residence time distribution method. Objective was to evaluate liquid holdup, axial dispersion and pressure drop of reactor bed consisting of catalyst particles diluted by inert fines to obtain data for process description by PD mathematical model. Hydrodynamic experiments were carried out in model glasses reactor with I.D 30.4 mm. Catalyst –trilobe extrudes of 1.3 mm O.D. was diluted by fine grain SiC 0.1-0.15 mm to obtained complete catalyst wetting and uniform liquid distribution. In presumed range of liquid and gas flow rates, air and water mass flow corresponded to values of hydrogen and hydrocarbons mixture in high-pressure reactor for the study of HDS catalysts. Gas flow was measured by MFC and liquid flow rate was checked by weighing, pressure drop was measured by pressure probe. Along the bed were located three electrodes to measure conductivity signal of responses to impulse of KCl solution. Data acquisition was made by four channel chromatography Clarity SW/HW, with subsequent evaluation by program in Matlab. Simultaneously, the reproducibility of three bed sections of trickle bed catalyst reactor formation of bed diluted by fines was tested by two methods. Effective mean residence time, liquid holdup and axial dispersion were evaluated for each. The range of experimental operation conditions (gas and liquid flow rates) negligible influenced by hydrodynamics was evaluated both experimentally and by hydrodynamics PD model

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1. Introduction

Trickle bed reactors are widely used for heterogeneously catalyzed reactions in liquid and solid phase or gas phase reaction on solid catalysts particle in inert liquid presence. Trickle bed reactors are industrially used in chemical and petrochemical processes of hydrogenation intermediates, hydrodesulphurization of petrochemical fractions, refining hydrogenation of alcohols, oxosynthesis products etc.

Laboratory scale reactors have several utilizations. They may be used as test reactors for development of new technology yet not carried out in industry scale, for intensification present technologies, for new catalyst application research, in case of change inlet reaction conditions and so on. Their use is essential in the study of long-therm catalyst deactivation.

In the ideal case, flowing liquid in trickle bed reactor can exhibit piston flow. At experimental conditions is often low liquid velocity used; therefore there is deviation from ideal flow presented, and thus the liquid maldistribution in bed causes, increasing wall flow, catalyst particle bypassing and local overheating of catalytic bed due to non-ideal heat transfer caused by liquid stagnant zones.

It is suitable to dilute catalyst bed with small innert particle to minimize this negative phenomenon. Presence of innert particles provides uniformity of liquid distribution and catalyst wetting. [1-3]

Hydrodynamic conditions in reactor are characterized especially by axial dispersion that occurs with the deviation from ideal piston flow, by liquid and gas holdup and pressure drop. Some of these parameters are possible to detect by residence time distribution of tracer liquid, when is investigated the character of liquid flow by catalyst bed by method impulse and response. [4-5]

The aim of this work was to compare the properties of diluted catalyst beds made by different packet methods. The task was to determine hydrodynamic characteristics (pressure drop and liquid holdup) and Peclet number for trickle downflow and flooded upflow reactor regime. The RTD method of KCl tracer was used to determine liquid holdup and axial mixing in the model of trickle bed reactor. Measured data were evaluated by PD model fit and compared with the results obtained by moment method.

Nomenclature

E	distribution function
F_G	gas flow rate (ml/min)
F_L	liquid flow rate (ml/min)
G	superficial gas flow rate ($kg.m^{-2}s^{-1}$)
GHSV	gas hourly space velocity (h^{-1})
h	liquid holdup
L	superficial liquid flow rate ($kg.m^{-2}s^{-1}$)
LHSV	liquid hourly space velocity (h^{-1})
Pe	Peclet number
tm	residence time (s)

2. Experimental setup

2.1. Upflow/downflow reactor

Hydrodynamics model of trickle bed test reactor consisting of glass tube of internal diameter 30.4 mm and total length 765 mm. There were placed three electrodes axially, in the upper, middle and bottom of the inside for measuring tracer conductivity. Axially placed common electrode diameter was 0.8 mm. Reactor was filled by trilobe catalyst diluted by innert particles SiC. Two different packet methods were used. Water and air was introduced by centrally located jet. Tracer liquid pulse was dosed by loop volume of 52 µl, conductivity change was measured by three conductometers OK 104 with ranges: electrode 1: 300 $\mu S.cm^{-1}$, electrode 2: 3 $mS.cm^{-1}$ and electrode 3: 30 $mS.cm^{-1}$.

Water was dosed into reactor continually by HPLC pump, air was supplied continuously throught mass flowmeter (FMA 1900 Omega Engineering) and pressure drop indicates by sensor (BD Sensor) on the top of reactor near liquid distributor.

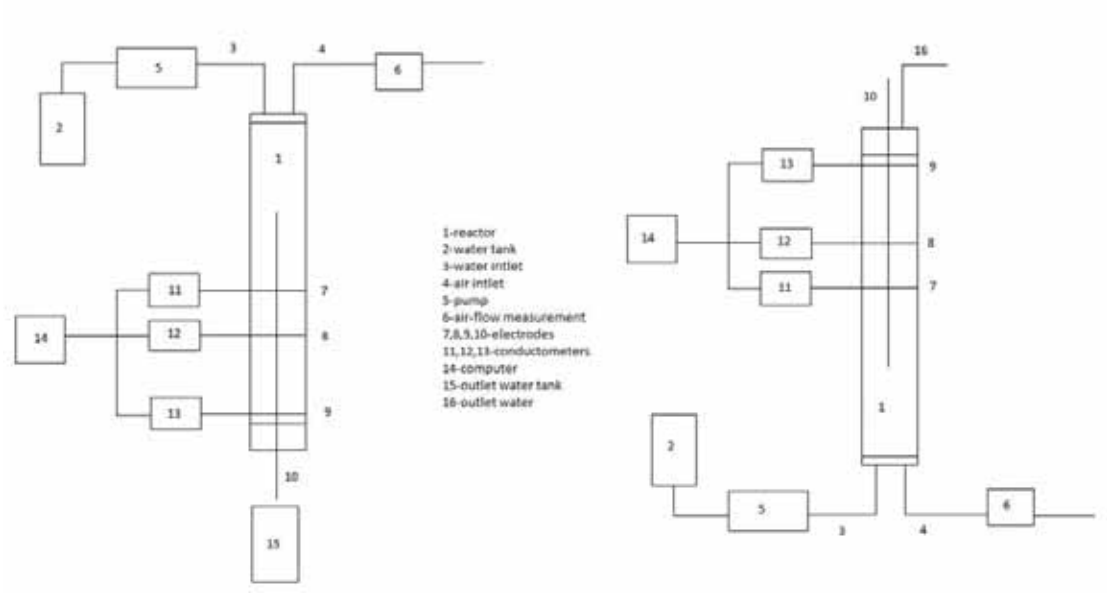

Fig. 1. Reactors scheme both for (a) down and (b) upflow regimes

Table 1: Experiments conditions

F_L(ml/min)	L ($kg.m^{-2}s^{-1}$)	LHSV (h^{-1})	F_G(ml/min)	GHSV(h^{-1})	G ($kg.m^{-2}s^{-1}$)
5.0	0.1234	3.5059	142	85.2	4.20E-03
3.3	0.0814	2.3373	95	57.0	2.81E-03
1.7	0.0419	1.1686	47	28.2	1.39E-03
0.8	0.0197	0.5843	24	14.4	7.11E-04

Range of liquid and gas superficial velocities is presented in table 1.

Reactor allowed measurement in both regimes; trickle and flooded bed bubbling regime. Reactor was modified to upflow so that bed and electrodes setting depending on direction flow was the same as in downflow regime. Three types of measurement were carried out: without catalytic bed with water and/or air injection, and two phase flow measurement in catalytic bed. The air and water flow rates were the same as in case of trickle regime.

2.2. Catalytic bed

Catalytic bed was divided into three sections with different composition. First section at the top consisted of 1-2 mm SiC and provided uniform liquid distribution. Second section was consisted of 0.1-0.2mm SiC and catalyst in volumetric ratio 2:1, third section consisted of 0.1-0.2mm SiC and catalyst in volumetric ratio 1:1 in case of trickle bed.

First section was the same in case of bubble flow, second and third sections were different. The volumetric ratio was conserved, instead 0.1-0.2 mm SiC was used 1-2 mm SiC a like in first section.

The dry packing method was used. Portion of catalyst and inert were mixed in beaker in the relevant ratio and packed into the reactor in several doses and tapped.

The boundaries of separate sections were indicated by electrode placing.

Measured data was recorded by Clarity chromatography SW and evaluated by Matlab program. The axially dispersed piston flow model characterized by dimensionless equation was used for evaluation.

Parameters of equation [6] were evaluated for closed-open boundary conditions provided relationship for system impulse response:

$$E(\Theta) = \frac{1}{2}\left(\frac{Pe}{\pi\Theta}\right)^{1/2} \exp\left(-\frac{Pe(1-\Theta)^2}{4\Theta}\right) - \frac{Pe}{2} e^{Pe} erfc\left[\frac{(Pe/\Theta)^{1/2}(1+\Theta)}{2}\right] \quad (1)$$

where Θ is dimensionless time t/t_m and Pe, Peclet number.

As an initial value for Nelder-Mead data fitting values evaluated by moment method were used.

Parameters of equation (1) (Pe and tm) were identified by simplex method in Matlab using the method of the least squares.

3. Results and discussion

Trickle bed reactor hydrodynamic conditions have influenced both catalyst and inert particle sizes, and composition of catalytic bed. Liquid holdup in bed increased with presence of fines fraction of SiC and its volumetric ratio due to catalyst particle (Fig. 2 a). SiC particles size has an influence on axial dispersion too. Rougher filler leads to larger deviation from the desired piston flow. Liquid and gas flow rate has no influence on axial mixing in the first section filled by SiC fines. On the other hand, its influence is significant for the remaining two sections composed from mixture of catalyst extrudates and diluent particles. Primarily when used at the lowest air and water flow rates (24 ml/min and 0.8 ml/min) , it leads to significant decrease of Peclet number in cases of used 0.1-0.2 mm SiC too.

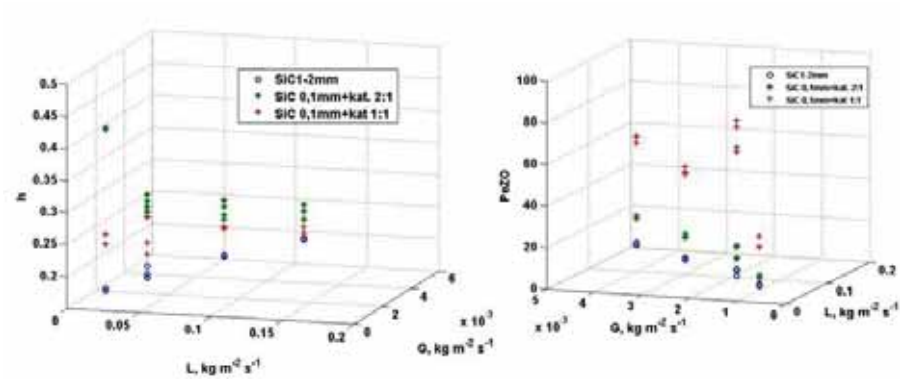

Fig. 2. (a) Influence of gas and liquid flow rates on liquid holdup, (b) Dependence of Peclet number closed-open boundary conditions on liquid and gas flow rates

Reactor pressure drop exhibits linear dependence proportional to flow rates (Fig. 3 a).

Residence time in reactor does not depend on filler particle size too much, however depends more on gas and liquid flow rates. With increasing flows residence time decreases rapidly.

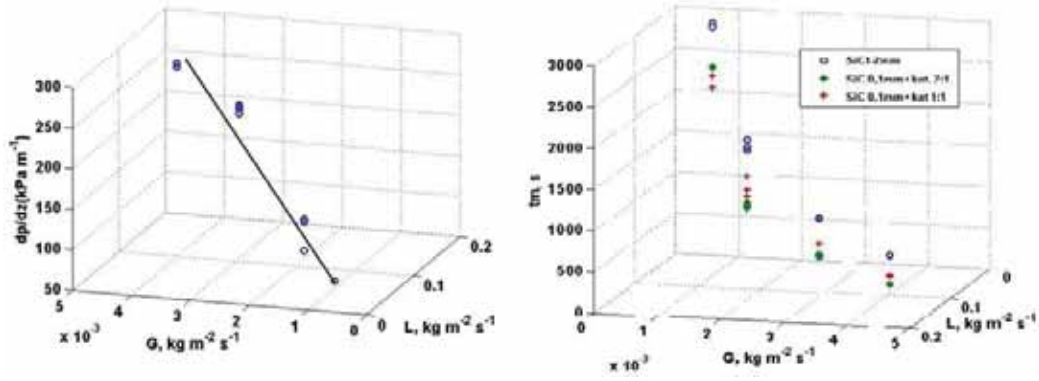

Fig. 3. (a) Dependence of pressure drop on liquid and gas flow rates, (b) Residence time in relation to gas and liquid flow

While measuring up flow bubble regime without padding, considerable signal oscillation occurred due to the presence of large air bubbles (Fig. 4). Distribution function on Fig. 4a corresponds to typical one mixer response. Experiments without air presence were carried out with three largest liquid flows only, because flows smaller than 5 ml/min provided too long residence time due to the extensive signal tailing. Experiments under these conditions were not feasible.

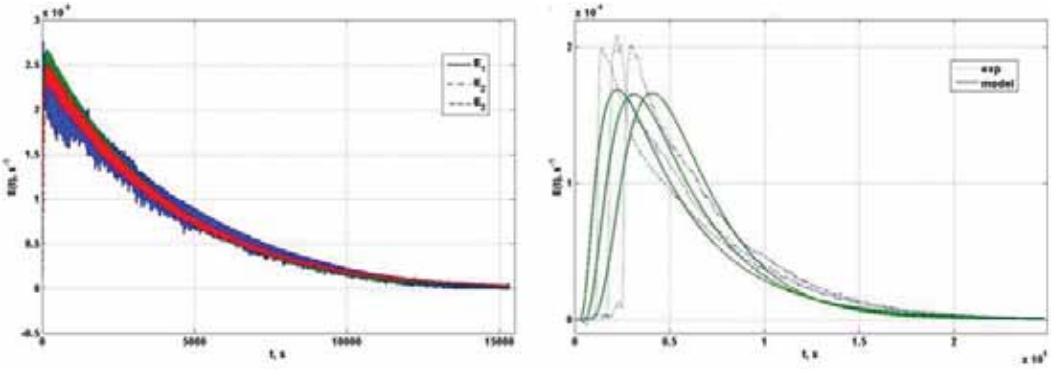

Fig. 4. Measurement of flooded reactor without packing, (a) inlet water-air (L=8.3 ml/min, G=234ml/min), (b) inlet water only (L=8.3 ml/min)

In terms of arrangement, axial mixing influence is stronger in case of flooded bed regime. Probably, the porosity of catalytic particle has insignificant influence on deviation from piston flow. As a consequence of this, outlet signal is strongly tailing and is unsuitable for PD model application. Peclet number is two-times smaller than in case of the trickle regime. Compared to trickle regime corresponded with PD model better. (Fig. 5). Residence time in flooded bed is two-times longer than in trickle layout in the same flows, which results is higher axial mixing again. Pressure drop in upflow regime is constant, and is not affected by liquid flow. On gas flow depends very few.

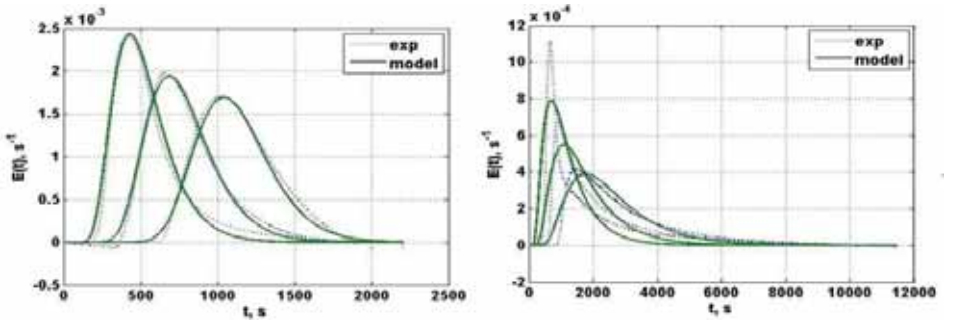

Fig. 5. (a) trickle flow, correspondence with PD, intlet section 1-2mm SiC Pe=6.2, middle section 0.1-0.2mm SiC + catalyst in ratio 2:1 Pe=21.2 and last section 0.1-0.2 mm SiC + catalyst in ratio 1:1 Pe= 13.8; (b) bubble flow different from PD model, intlet section 1-2mm SiC Pe=8.5, middle section 1-2mm SiC + catalyst in ratio 2:1 Pe=4.1 and last section 1-2 mm SiC + catalyst in ratio 1:1 Pe= 13.5

4. Conclusion

Trickle bed reactor behavior is significantly influenced both by bed structure and composition. Depending on catalysts bed structure, it can cause undesirable phenomena as non-uniformity wetting of catalytic particles, wall flow and axial dispersion increasing due to deviation from piston flow. To prevent these imperfections it is recommended to use adequately small catalytic particles considering reactor diameter, and diluted catalytic bed by small inert particles, with idealize flow.

Hydrodynamic conditions in diluted catalytic bed were observed by residence time distribution method. Measured characteristics were processed both by moment method and PD model.

Reactor was divided into three sections with different composition to observe particle size and structure influence. An intense influence of axial dispersion and low liquid holdup was observed with largest particles SiC (1-2mm), on the other hand catalytic particles in combination with fine-grained SiC evince more ideal flow and better particle wetting.

The influence of liquid and gas flow rates were observed too. Liquid holdup and axial mixing increase were strong in case of trickle regime in lower flow rates.

Upflow regime provided significant tailing of signal response to impulse. Hydrodynamic results in case of axial mixing; however liquid holdup providing desirable catalyst particle wetting is about 80 percent.

Air and water flow rates has not influence on pressure drop of catalytic bed in upflow.

Reduction of air flow has significant influence on increasing residence time.

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