STUDIES IN MULTIPHASE REACTORS

By

ARUNABHA KUNDU

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Dedicated to

My Parents and Grandparents

CERTIFICATE

This is to certify that this thesis entitled "**Studies in Multiphase Reactors**" being submitted by **Mr. Arunabha Kundu** to the Indian Institute of Technology, Delhi for the award of the degree of Doctor of Philosophy is a record of bona fide research work carried out by him. Mr. Kundu has worked under my supervision and has fulfilled the requirement for the submission of the thesis. The results contained in this thesis have not been submitted in part or full, to any other university or institute for the award of any degree or diploma.

(Prof. K. D. P. Nigam)
Professor
Department of Chemical Engineering
Indian Institute of Technology, Delhi

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Abstract

Multiphase reactors are widely used in diverse applications such as in manufacture of petroleum-based fuels and products, in production of commodity and speciality chemicals, pharmaceuticals, herbicides and pesticides, in refining of ores, in production of polymers and other materials and pollution abatement. In all such applications, the knowledge of fluid dynamic parameters is necessary for development of appropriate reactor models and scale-up rules. These reactors can be operated in gas-liquid, gas-liquid-solid (solids are either fixed or suspended) and gas-liquid-liquid (having an immiscible second liquid phase). The design of multiphase reactors is complex due to the highly complex nature of hydrodynamic condition and the reaction mechanism. In the present work, studies on hydrodynamic parameters of two types of multiphase reactors are presented. Therefore the thesis has been divided into two parts viz.

1. Part A. **Gas-Liquid-Solid System in Trickle-Bed Reactors**
2. Part B. **Gas-Liquid-Liquid System in Bubble Column Reactors**

Part A

Trickle-bed reactors, in which liquid and gas flow concurrently downward through a packed catalyst bed, are common in the petrochemical industry. In refining crude oil, they are used to upgrade naphtha, distillate or heavy oil by treating with hydrogen over a catalyst to remove impurities and saturate aromatics. At reaction conditions, the petroleum streams are often a mixture of vapor and liquid or essentially all liquid fed concurrently with hydrogen gas.

The use of trickle-bed reactors (TBRs) in petroleum refining industries in the "More Miles with fewer Emissions" age finds spectacular growth. This century demands more quantity of very clean gasoline or diesel with tighter environmental protection regulations in

contrast to increased availability of heavy and sour crudes. Only trickle-bed reactor can cope up with the present situation. The increased importance of hydroprocessing has spurred research interest in the proper design and scale up of TBR .

Of the flow regimes identified in trickle beds, commercial hydrotreating units operate in either the trickling or pulsing flow. The trickling flow regime is observed at relatively small liquid flow rates and small to moderate gas flow rates. Gas flow is continuous, while the liquid forms continuous films or rivulets over the packing surfaces. The pulsing flow prevails over a large range of intermediate and high gas and liquid flow rates. It is characterized by the alternating passage of liquid-rich and gas-rich zones (pulses) and associated with high heat and mass transfer rates.

Liquid distribution in trickle-bed reactors is often not uniform, which has detrimental effects on reactor performance, such as loss of catalyst effectiveness, creation of hot spots, etc. One of the most important factors influencing maldistribution appears to be the liquid distribution at the top of the bed. The liquid distribution studies were carried out in a 0.0152 m diameter trickle-bed reactor with a uniform liquid distributor. Different liquids (water, kerosene, water+surfactant, water+ethylene glycol) were used with five different catalyst packing different in shape and size.

The main objective of this work was to develop a physical model describing the hydrodynamics of the cocurrent gas–liquid trickle flow through a trickle-bed reactor. The trickle flow regime was modelled by an annular flow in which the gas and liquid phases are separated by a smooth and stable interface. Attention was paid to the formulation of the model on the basis of the fundamental force balance equations applied to each phase flow. Furthermore, the formulation of the mutual interactions between the three phases was investigated. The predictions of the pressure gradient and liquid saturation were compared with experimental data of the literature obtained in a wide range of operating pressures. The model was further extended to predict the external wetting efficiency (which is defined as fraction of the external catalyst pellet area wetted by the flowing liquid) in trickle-bed reactors. The radioisotope tracer studies were also carried out and the experimental value of external wetting efficiency was obtained from the square root of the ratio of the effective diffusivity in two phase flow to the effective diffusivity in liquid filled bed under the same intrinsic velocity of liquid.

Part B

Gas-liquid-liquid systems are used in aerobic bioprocesses, biological waste water treatment and liquid phase heterogenisation in catalysis by coordination complexes (biphasic catalysis). Biphasic catalytic systems are used because of the easy separation of the catalysts from the organic products (as the catalyst is soluble in the aqueous phase only). In such media a significant increase of the gas-aqueous liquid mass transfer rate were observed (Lekhal et al., 1997). Mass transfer behaviour is important for the gas-liquid-liquid reaction system with fast parallel and consecutive reactions as its effect on the product yield can be significant (Brilman, 1998). For systems with slow reaction kinetics, it will not make any problem as the three phases will be at physical equilibrium.

The enhanced absorption rate may be caused by changes in the gas-liquid interfacial area a, through gas phase hold-up and mean bubble size variation, or by increasing the liquid side mass transfer coefficient, k_L. The existing literature reveals that the addition of an immiscible organic liquid phase to a gas–aqueous liquid phase system may have complex effects and may significantly influence oxygen transfer by absorbing oxygen or supplying it to other phases, or by influencing the fluid mechanics of the continuous phase, thereby influencing mass transfer coefficient and interfacial area of the continuous phase.

Though maximum existing literature have been reported on gas-liquid-liquid measurements performed in stirred vessels, specific information with G-L-L system in bubble columns is quite limited (Yoshida et al., 1970; Sharma and Mashelkar, 1968; Kato et al., 1985). The effects reported on mass transfer parameters, like gas-liquid interfacial area and gas hold-up, were discussed shortly. No general trends can be derived from the reported results in the literature.

Experimental results of mass transfer in air-water system with a dispersed immiscible organic liquid in a bubble column 0.076 m in diameter for seven different organic liquids at various hold-up are presented. Experiments were carried out at gas superficial velocity in the range 0.0052-0.026 m/s at 293K. The volumetric mass transfer coefficient, k_La was determined by dynamic gas absorption technique. Overall gas hold-up was also measured.

Contents

		Page Number
Certificate		iii
Acknowledgements		iv
Abstract		v-viii
Contents		ix-xii
List of Figures		xiii-xx
List of Tables		xxi

Part A: Gas-Liquid-Solid System in Trickle-Bed Reactors

Nomenclature		2-4
1. Chapter I	**Literature Review**	5-32
1.1	**Introduction**	6-9
1.2	**Hydrodynamic parameters**	9-32
1.2.1	**Pressure Gradient and Liquid Hold-up**	11-16
1.2.2	**Flow Regime**	16-22
1.2.3	**Liquid Distribution**	22-29
1.2.4	**Wetting Efficiency**	29-32
2. Chapter II	**Experimental Section**	33-42
2.1	**Experimental Set-Up**	34-40
2.2	**Startup Procedure**	40-42

3. Chapter III	**Development of Hydrodynamic Models for Gas-Liquid-Solid Reactors**	43-55
3.1	**Modeling**	44-46
3.1.1	Gas Phase Force Balance	45
3.1.2	Liquid Phase Force Balance	45-46
3.2	**Modeling of the Drag Forces**	46-49
3.2.1	Particle-Gas Drag Model	46-47
3.2.2	Particle-Liquid Drag Model	47
3.2.3	Liquid-Gas Interfacial Drag Model	48-49
3.3	**Model for Tortuosity Effect**	49-52
3. 4	**Combination of the Models**	52
3.5	**Model for Wetting Efficiency**	52-55
3.5.1	Strategy for the estimation of the parameters	53-55
4. Chapter IV	**Results and Discussion**	56-87
4.1	**Results: Liquid Distribution**	57-60
4.1.1	Effect of properties of liquid	57
4.1.2	Effect of liquid flow rate	58
4.1.3	Effect of Gas flow rate	58-60
4.1.4	Effect of Particle Geometry	60
4.2	**Results: pressure-drop and liquid hold-up**	60-68
4.2.1	Comparison of predictions from model and available correlations with experimental data	67-68
4.3	**Results: Wetting Efficiency**	69-87

5. Chapter V	**Conclusion**	88-90
5.1	**Conclusion**	89-90
5.2	**Scope of Future Work**	90
References		91-104
Appendix I: Method of Calculation		105-107
Appendix II: Mathematical Modeling of Flow Regimes		108
Appendix III: Computer program Listing		109-121

Part B. Gas-Liquid-Liquid System in Bubble Column Reactors

Nomenclature		123-125
1. Chapter I	**Literature Review**	126-161
1.1	**Introduction**	127-131
1.2	**Observations in Gas-Liquid-Liquid Systems**	131-132
1.3	**Parameters affected in the presence of the dispersed liquid phase**	132-148
1.3.1	**Mass transfer in the boundary layer: possible pathways**	133-141
1.3.2	**Effect of oil addition on k_La**	141-145
1.3.3	**Effect of oil addition on gas–liquid interfacial area 'a'**	145-148
1.4	**Mass transfer enhancement**	148-160
1.4.1	**Enhancement factor definition**	148-149
1.4.2	**"Bubble covering" mechanism**	149-152
1.4.3	**"Shuttle effect" mechanism**	153-155

1.4.4	"Permeability effect"	155-160
1.5	Concluding Remarks	160-161
2. Chapter II	**Experimental Section**	162-174
2.1	Experimental Set-Up and procedure	163-166
2.2	Theory for evaluation of volumetric mass transfer coefficient, k_La	167-174
2.2.1	Derivation of k_La	167-173
2.2.2	Calculation of Enhancement Factor (E)	173-174
3. Chapter III	**Results and Discussion**	175-188
3.1	Results and Discussion	176-188
3.1.1	General Observations	176-183
3.1.2	Influence of u_g and ϕ_{disp} on k_La	183-188
4. Chapter IV	**Conclusion**	189-191
4.1	Conclusion	190-191
References		192-197
About the Author		198-199