

**BED STRUCTURE AND ITS IMPACT ON HYDRODYNAMICS  
OF TRICKLE BED REACTORS**

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**BED STRUCTURE AND ITS IMPACT ON HYDRODYNAMICS  
OF TRICKLE BED REACTORS**

*by*

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*Submitted*

*in fulfillment of the requirements of the degree of Doctor of Philosophy*

*to the*



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## **CERTIFICATE**

This is to certify that the thesis entitled '**Bed Structure and Its Impact on Hydrodynamics of Trickle Bed Reactors**' being submitted by **Ms. Akarsha Srivastava** to the Indian Institute of Technology Delhi for the award of degree of **Doctor of Philosophy** is a record of bona fide research work carried out by her under our guidance and supervision. The research report and results presented 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.

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Akarsha Srivastava

## ABSTRACT

Various petroleum refining and petrochemical industries use Trickle Bed Reactors (TBR) to treat the petroleum feedstock with hydrogen to remove sulfur (hydrodesulfurization (HDS)), nitrogen (hydrodenitrification (HDN)), oxygen (hydrodeoxygenation (HDO)), metal components (hydrodemetallization (HDM)), and to saturate aromatic rings (hydrodearomatization (HDA)) etc. at high temperatures and pressures. Trickle Bed Reactors are multiphase packed bed reactors (Gas-Liquid-Solid) with gas and liquid both flowing co-currently downward through the bed of catalyst particles. These catalyst particles (on the internal surface of which the desired reaction takes place) are charged into the reactor in a grossly random fashion, resulting in a heterogeneous or non-uniform structure of the bed.

In reactions where “deep conversion” is desired, local non-uniformities in packing structure in a TBR is thought to have a dramatic impact on its performance. With the environmental laws and imposition of stricter limits for sulfur in ULSD (Ultra Low Sulphur Diesel) at extremely low levels of 10 and 15 ppm, the conversion levels or the performance of TBR’s need to be upgraded to higher levels. These bed defects, stated earlier, tend to create non-uniform voidage variation inside the bed. Local variation in voidage in turns affects the flow related parameters, such as liquid-gas distribution, axial dispersion, wetting and irrigation of pellets, formation of hot spots, etc., and in turn adversely affect reactor performance and catalyst life.

Structure of packed bed is composed of many factors such as filling pattern, particle size, and shape, column-particle-diameter ratio and particle size distribution. The aim of this investigation is to characterize the packing structure of randomly packed beds of monosized spherical particles. For extracting this quantitative information, an extensive characterization method has been used in this work in which structure of dense-packed bed has been generated

experimentally, its three-dimensional structure experimentally determined using bed freezing, sectioning and image analysis. With this exact information on the location of particles, their size and packing fraction, statistical measurements such as radial variations in voidage, radial distribution function and nearest neighbour function based on the interparticle distances have been determined.

Packing heterogeneity and its effect on the flow properties of the resulting packed bed is studied in a quasi-two-dimensional column as it helps in visualizing the structure which is not easily amenable in a curved geometry. The packing patterns are first generated by an automated set-up to ensure reproducibility and their natural packing structure is further examined so that it can be individualized. Moreover, fluid flow behavior in various packing arrangements is studied.

Uniform liquid distribution is crucial in Trickle Bed Reactors for the proper functioning of the reactor. Poor distribution is attributed to the poor design of distributor, wall flow at the bed scale and voidage variation at particle scale caused either by the particle shape or by the pore space geometry. With packing defects, further study is required to understand the flow behavior of fluids inside the bed. The fluid flow pattern resulting from different loading techniques (discussed earlier) is mapped by keeping other factors (particle shape and column-particle-diameter ratio) constant. The impact of other deciding factors such as aspect ratio of the column, prewetting, and fluid flow rates on liquid flow behavior has been examined.

## सार

विभिन्न पेट्रोलियम रिफाइनिंग और पेट्रोकेमिकल उद्योग सल्फर (हाइड्रोडेसल्फ्यूरेशन (एचडीएस)), नाइट्रोजन (हाइड्रोडेनेट्रीफिकेशन (एचडीएन)), ऑक्सीजन (हाइड्रोडेक्सीकसीनीकरण (एचडीओ)), धातु घटकों (हाइड्रोडेमेटलाइजेशन (एचडीएम)) को हटाने के लिए हाइड्रोजन के साथ पेट्रोलियम फीडस्टॉक का इलाज करने के लिए और उच्च तापमान और दबाव पर हाइड्रोडाइएरोमेटाइजेशन (HDA) को संतृप्त करने के लिए ट्रिकल बेड रिएक्टर (टीबीआर) का उपयोग करते हैं। ट्रिकल बेड रिएक्टर गैस और तरल के साथ पैक किए गए (गैस-लिक्विड-सॉलिड) मल्टीफेज रिएक्टर हैं, जो उत्प्रेरक कणों के पैकिंग के माध्यम से वर्तमान में नीचे की ओर बहते हैं। इन उत्प्रेरक कणों (जिस पर वांछित आंतरिक प्रतिक्रिया होती है) को रिएक्टर में स्थूल रूप से बेतरतीब ढंग से चार्ज किया जाता है, जिसके परिणामस्वरूप पैकिंग की विषम या गैर-समान संरचना होती है।

प्रतिक्रियाओं में जहां "गहरा रूपांतरण" वांछित है, टीबीआर में पैकिंग संरचना में स्थानीय गैर-एकरूपता के बारे में सोचा जाता है कि इसके प्रदर्शन पर गंभीर प्रभाव पड़ता है। ULSD (अल्ट्रा लो सल्फर डीजल) में सल्फर के लिए पर्यावरणीय कानूनों और सख्त सीमाओं को लागू करने के साथ, 10 और 15 पीपीएम के निम्न स्तर पर, रूपांतरण स्तर या TBR के प्रदर्शन को उच्च स्तर पर अपग्रेड करने की आवश्यकता है। ये पैकिंग के दोष, जो पहले कहा गया था, पैकिंग के अंदर गैर-समान रूप से भिन्नता पैदा करते हैं। शून्यता (Voidage) में स्थानीय भिन्नता प्रवाह से संबंधित मापदंडों को प्रभावित करती है, जैसे कि तरल-गैस वितरण, अक्षीय फैलाव, उत्प्रेरक कणों को गीला करना और सिंचाई करना, गर्म स्थानों का निर्माण, आदि, और बदले में रिएक्टर प्रदर्शन और उत्प्रेरक जीवन को प्रतिकूल रूप से प्रभावित करते हैं।

पैकड बेड की संरचना कई कारकों से बनी होती है जैसे कि पैकिंग पैटर्न, कण आकृति और आकार, स्तंभ-कण-व्यास अनुपात और कण आकार वितरण। इस जांच का उद्देश्य मोनोसाइज्ड गोलाकार कणों के

बेतरतीब ढंग से पैक बेड की पैकिंग संरचना की विशेषता है। इस मात्रात्मक जानकारी को निकालने के लिए, इस काम में एक व्यापक लक्षण वर्णन पद्धति का उपयोग किया गया है जिसमें घने-पैकिंग की संरचना को प्रयोगात्मक रूप से उत्पन्न किया गया है, इसकी तीन आयामी संरचना प्रयोगात्मक रूप से बेड फ्रीजिंग, सेक्शनिंग और छवि विश्लेषण का उपयोग करके निर्धारित की गई है। कणों के स्थान, उनके आकार और पैकिंग अंश पर सटीक जानकारी के साथ, सांख्यिकीय माप जैसे कि शून्य (voidage) में भिन्नता, रेडियल वितरण फ़ंक्शन और इंटरपेरिकल दूरी के आधार पर निकटतम पड़ोसी फ़ंक्शन का निर्धारण किया गया है।

पैकिंग विषमता और इसके परिणामस्वरूप पैक किए गए पैकिंग के प्रवाह गुणों पर इसका प्रभाव एक अर्ध-द्वि-आयामी कॉलम में अध्ययन किया जाता है क्योंकि यह संरचना को देखने में मदद करता है जो घुमावदार ज्यामिति में आसानी से प्राप्य नहीं है। पैकिंग पैटर्न सबसे पहले एक स्वचालित सेट-अप द्वारा निर्मित किया जाता है ताकि प्रजनन और उनकी प्राकृतिक पैकिंग संरचना की जांच की जा सके ताकि इसे अलग-अलग किया जा सके। इसके अलावा, विभिन्न पैकिंग व्यवस्था में द्रव प्रवाह व्यवहार का अध्ययन किया गया है।

रिएक्टर के समुचित कार्य के लिए ट्रिकल बेड रिएक्टरों में समान तरल वितरण महत्वपूर्ण है। खराब वितरण को वितरक के खराब डिजाइन, पैकिंग पर दीवार के प्रवाह और कण आकार या कण ज्यामिति पर शून्य भिन्नता के लिए जिम्मेदार ठहराया जाता है। पैकिंग दोषों के साथ, पैकिंग के अंदर तरल पदार्थ के प्रवाह व्यवहार को समझने के लिए आगे के अध्ययन की आवश्यकता होती है। विभिन्न लोडिंग तकनीकों (पहले चर्चा की गई) से उत्पन्न द्रव प्रवाह पैटर्न अन्य कारकों (कण आकार और स्तंभ-कण-व्यास अनुपात) को स्थिर रखते हुए मैप किया गया है। अन्य निर्णय लेने वाले कारकों जैसे स्तंभ के आस्पेक्ट अनुपात, प्रीवेटिंग, और तरल प्रवाह व्यवहार पर द्रव प्रवाह दर की जांच की गई है।

# TABLE OF CONTENTS

<b>CERTIFICATE</b>	
<b>ACKNOWLEDGEMENTS</b>	
<b>ABSTRACT</b>	<b>(i)</b>
<b>TABLE OF CONTENTS</b>	<b>(iii)</b>
<b>LIST OF FIGURES</b>	<b>(vii)</b>
<b>LIST OF TABLES</b>	<b>(xiii)</b>
<b>1. Introduction, Motivation, and Objectives</b>	<b>1</b>
1.1 Overview	1
1.2 Motivation	6
1.3 Objectives	12
1.4 Structure of the thesis	13
Notation	15
References	16
<b>2. Literature Review</b>	<b>19</b>
2.1 Packing of particles in TBR	19
2.1.1 Packing fraction/bulk voidage	25
2.1.2 Radial voidage	32
2.2 Hydrodynamics in TBR	48
2.2.1 Flow regimes	48
2.2.1 Two-phase pressure drop and liquid holdup	49
2.2.1 Mass transfer coefficients	58
2.2.1 Liquid distribution and wetting efficiency in TBR	60
2.3 Numerical models for Trickle Bed Reactor	68
2.4 Gaps in Literature	71
Notation	72
References	74
<b>3. Characterization of packing structure of mono-disperse spheres in cylindrical column</b>	<b>85</b>
3.1 Introduction	85

3.2	Experimental methodology	88
3.2.1	Packing experiments	88
3.2.2	Bed solidification, sectioning and image acquisition	91
3.2.3	Image analysis	91
3.3	Results and discussion	92
3.3.1	Effect of column-particle-diameter ratio	92
3.3.1.1	Axial and radial variation of voidage	93
3.3.1.2	Nearest neighbor distance distribution function	101
3.3.1.3	Radial distribution function	104
3.3.2	Effect of spatial arrangement of point processes	105
3.4	Conclusions	111
	Notation	112
	References	114
<b>4</b>	<b>Quantification of local structure of disordered packing of spherical particles</b>	<b>119</b>
4.1	Introduction	119
4.2	Experimental methodology	122
4.2.1	Packing Experiments	126
4.3	Theory: analysis of packing structure	129
4.4	Results and discussion	132
4.4.1	Overall voidage and radial voidage profiles	133
4.4.2	Radial distribution function	141
4.4.3	Local degree of disorder	142
4.5	Conclusions	149
	Notation	150
	References	152
<b>5.</b>	<b>Bed structure and its impact on liquid distribution in a Trickle Bed Reactor</b>	<b>156</b>
5.1	Introduction	156
5.2	Experimental methodology	160
5.3	Results and discussion	164

5.3.1	Two-phase pressure drop	164
5.3.2	Dynamic liquid holdup	167
5.3.3	Distribution index and flow maps	169
5.4	Conclusions	177
	Notation	179
	References	180
<b>6.</b>	<b>Investigation of packing patterns in a quasi-two-dimensional column</b>	<b>184</b>
6.1	Introduction	184
6.2	Experimental methodology	185
6.2.1	Generation of packing defects	186
6.2.2	Measurement of flow distribution in quasi-two-dimensional column	188
6.3	Results and discussion	191
6.3.1	Analysis of structure through imaging technique: Angle of Repose	191
6.3.2	Mean Dispersion	192
6.3.3.	Analysis of structure through solidification by resin: Kernel Density Estimation	198
6.3.4.	Measurement of flow distribution corresponding to the packing patterns	200
6.4	Conclusions	203
	Notation	203
	References	205
<b>7.</b>	<b>Conclusions and Recommendations for Future Research</b>	<b>207</b>
7.1	Summary of the present work	207
7.1.1	Characterization of packing structure of monodisperse spheres in a cylindrical column	208
7.1.2	Quantification of the local structure of disordered packing of spherical particles	209
7.1.3	Bed structure and its impact on liquid distribution in a trickle bed reactor	209

7.1.4	Investigation of packing patterns in a quasi-two-dimensional column	210
7.2	Thesis Accomplishments	211
7.3	Recommendation for future research	212
	BIO-DATA	

## LIST OF FIGURES

<b>Figure No.</b>	<b>Title</b>	<b>Page No.</b>
1.1	Schematic of a Trickle Bed Reactor (Mederos et al., 2009)	3
1.2	Image of packing in case of (a) Sock loading (b) Dense loading (U.S. Pat. No. 4,972,884)	7
1.3	Packing non-uniformities in Trickle Bed Reactor (a) Hollow (b) Donut (c) Bump (d) Slope	7
1.4.	Impact of loading pattern on liquid distribution (Petroval dense loading, <a href="https://www.petroval.com/densicat.html">https://www.petroval.com/densicat.html</a> )	8
1.5.	Effect of structure on design parameters of Trickle Bed Reactor	10
1.6.	Spatial variation of conversion in a transverse section of packed bed captured by volume selective spectroscopy reported by Yuen et al. (2002)	12
2.1	X-ray tomography images of packing structure of equilateral cylinders (a) three-dimensional view of aluminum core particles (b) two-dimensional image of packing of the equilateral cylinder (Zhang et al., 2006b)	21
2.2	The four platonic solids: tetrahedron (P1), icosahedron (P2), dodecahedron (P3), octahedron (P4)	22
2.3	Concentration profiles for the three-phase catalytic reaction in TBR (Ramachandran and Chaudhari, 1983)	58
2.3	(1) Two-dimensional packed bed with gas and liquid concurrent downflow and no-slip wall boundaries; (2) interconnected cell network; (3) Fluid superficial velocities and concentrations of species i at the interior face of the cell j. (Jiang et al., 2005)	70
3.1	Experimental methodology adopted for packing structure characterization	90
3.2	(a) Axial variation of voidage for $D_v/d_p = 14.28, 25$ and $66.67$ (b) Bottom end effects (c) Top end effect	94

<b>3.3</b>	(a) Sample image for $D_t/d_p = 66.67$ , (b) Sample image for $D_t/d_p = 25$ , (c) Sample image for $D_t/d_p = 14.28$ , (d) average image after superimposing all images for $D_t/d_p = 66.67$ , (e) $D_t/d_p = 25$ , (f) $D_t/d_p = 14.28$	95
<b>3.4</b>	Radial porosity variation for low range of $D_t/d_p$ ratio (a) $D_t/d_p = 4.0$ and (b) $D_t/d_p = 8.3$	96
<b>3.5</b>	Radial porosity variation for (a) $D_t/d_p = 14.3$ , (b) $D_t/d_p = 25.0$ and (c) $D_t/d_p = 33.3$	97
<b>3.6</b>	Radial porosity variation for (a) $D_t/d_p = 50.0$ , (b) $D_t/d_p = 66.7$ and (c) $D_t/d_p = 100.0$	98
<b>3.7</b>	Map for demarcation of low, intermediate and high range of column-particle-diameter ratio along with the existing wall, transition and core region (a) low range of $D_t/d_p$ (b) inntermediate range of $D_t/d_p$ and (c) high range of $D_t/d_p$	101
<b>3.8</b>	(a) Nearest neighbor function for variation in column-particle-diameter ratio; (b) First, second and third nearest neighbor function for column-particle-diameter ratio of 66.67, (c) for column-particle-diameter ratio of 25, (d) for column-particle-diameter ratio of 14.28	103
<b>3.9</b>	Radial distribution function for variation in column-particle-diameter ratio	105
<b>3.10</b>	Spatial point patterns with increasing degree of regularity	106
<b>3.11</b>	(a) Nearest neighbor function for point pattern 1, 2 ,3 and 4; (b) Nearest neighbor function for random, disordered and ordered packing; (c) Nearest neighbor distance and corresponding number of particles for ordered and disordered point patterns	108
<b>3.12</b>	Radial distribution function for point pattern 1, 2 ,3 and 4	109
<b>3.13</b>	Radial distribution function for random, disordered and ordered packing	110
<b>4.1</b>	Conical hopper set-up (a) schematic of packing set-up, (b) front view of packing set-up, (c) Insert-1 for Solid-Cone packing, (d) Insert-2 for Hollow-Cone packing, (e) image of conical hopper, (f) image of inserts for conical hopper (All dimensions are in mm)	124
<b>4.2</b>	Wedge-shaped hopper set-up (a) schematic of packing set-up, (b) top view of packing set-up, (c) front view of packing set-up, (d) image of packing set-up,	125

	(e) image of insert for Multi-Feed packing, (f) image of knife edge gate valve (All dimensions are in mm)	
<b>4.3</b>	Schematic for the six packing techniques (a) Vib-Fixed (b) Solid-Cone (c) Multi-Feed (d) Hollow-Cone (e) Central-Single-Source (f) Peripheral-Single-Source	126
<b>4.4</b>	Dirichlet Tessellation of Point Pattern	132
<b>4.5</b>	(a) Radial voidage for all packing methods along with Mueller's model prediction (b) Radial voidage profile near wall region	135
<b>4.6</b>	Comparison of radial voidage predictions by different correlations available in literature	136
<b>4.7</b>	Radial voidage along with proposed equation for (a) Vib-Fixed (b) Solid-Cone (c) Multi-Feed (d) Hollow-Cone (e) Central-Single-Source (f) Peripheral-Single-Source	140
<b>4.8</b>	Radial Distribution Function for packing methods	141
<b>4.9</b>	Protocol for quantification of local structure	143
<b>4.10</b>	Comparison of probability distribution functions along with the experimental area distributions	144
<b>4.11</b>	Area distribution for all packing methods at (a) $H/d_p=1$ , (b) $H/d_p=57$ , (c) $H/d_p=114$ , (d) $H/d_p=227$ , (e) $H/d_p=284$ , (f) $H/d_p=333$	146
<b>4.12</b>	(a) Axial profiles of mean for area distributions of all packing methods (b) coefficient of variation for area distributions of all packing methods	148
<b>5.1</b>	Different flow structures occurring in a trickle bed reactor, formation of film, rivulets, pendular structures, liquid pockets, and filaments (adapted from Lutran et al., 1991)	157
<b>5.2</b>	Schematic of automated experimental set-up for liquid distribution measurement	161
<b>5.3</b>	Set-up for liquid distribution collection technique employed in this study (i) Distinct features of set-up: 1- Distributor – spray nozzle, 2- Packed column, 3- Collecting device, 4- Manometers, 5- Sliding tray, 6- Collecting tray (ii) Collecting device along with silicone pipes for each collecting zone (iii)	163

	Isometric view of collector device (iv) Top view of collector device showing the collecting zones (v) Dimensions of collector device	
<b>5.4</b>	Two phase pressure drop for all packing methods at (a) $L = 3 \text{ kg/m}^2.\text{s}$ , (b) $L = 8.5 \text{ kg/m}^2.\text{s}$ , (c) $L = 12.7 \text{ kg/m}^2.\text{s}$	166
<b>5.5</b>	(a) Parity plot of experimental and predicted pressure drop (b) Pressure drop data for PM-1 by experiments and predicted by correlation	167
<b>5.6</b>	Dynamic liquid hold-up for all packing methods at (a) $L = 3 \text{ kg/m}^2.\text{s}$ , (b) $L = 8.5 \text{ kg/m}^2.\text{s}$ , (c) $L = 12.7 \text{ kg/m}^2.\text{s}$	169
<b>5.7</b>	(a) Parity plot of experimental and predicted hold-up (b) Hold-up data for PM-1 by experiments and predicted by correlation	169
<b>5.8</b>	Maldistribution index for all packing methods for liquid flow range $L = 3 \text{ kg/m}^2.\text{s} - 12.7 \text{ kg/m}^2.\text{s}$	170
<b>5.9</b>	Flow map for Vib-Fixed (i) $L = 3 \text{ kg/m}^2.\text{s}$ , (ii) $L = 8.5 \text{ kg/m}^2.\text{s}$ , (iii) $L = 12.7 \text{ kg/m}^2.\text{s}$	171
<b>5.10</b>	Maldistribution index for all packing methods in trickle flow regime for prewetted conditions; $L/D = 5$ , $L = 3 \text{ kg/m}^2.\text{s}$	172
<b>5.11</b>	Maldistribution index for all packing methods for different column aspect ratios $L/D = 1/8, 1/4, 1/2, 1, 2, 3, 4, 5$	172
<b>5.12</b>	Flow map of all packing methods at $L = 3 \text{ kg/m}^2.\text{s}$ and $G = 0.078 \text{ kg/m}^2.\text{s}$ for pre-wetted packed bed (A) Vib-Fixed (B) Solid-Cone (C) Multi-Feed (D) Hollow-Cone (E) Central-Single-Source (F) Peripheral-Single-Source for $L/D=0.5$ (i), 2 (ii), 5 (iii)	176
<b>5.13</b>	Maldistribution index for different types of distributors for a uniform packing (Vib-Fixed), $L = 3 \text{ kg/m}^2.\text{s}$ and $G = 0.078 \text{ kg/m}^2.\text{s}$	176
<b>5.14</b>	Maldistribution index for different types of distributors for a non-uniform packing, $L = 3 \text{ kg/m}^2.\text{s}$ and $G = 0.078 \text{ kg/m}^2.\text{s}$	177
<b>6.1</b>	(a) Schematic of packing setup (all dimensions are in mm) (b) Image of front view and side view of laboratory packing set-up	187
<b>6.2</b>	Images of the packing defects along with the angle of repose as determined by Image Processing for (a) Hollow (b) Donut (c) Bump (d) Slope	188
<b>6.3</b>	Experimental set-up manufactured for studying liquid flow distribution	190

<b>6.4</b>	Different packed structures as a result of intermittent flow in the hopper (a) Packing-1 (b) Packing-2 (c) Packing-3 (d) Packing-4	193
<b>6.5</b>	Different packed structures as a result of continuous flow of stacked particles in the hopper (a) Packing-1 (b) Packing-2 (c) Packing-3 (d) Packing-4	194
<b>6.6</b>	Image of the packing structure after the background has been subtracted from the whole image to determine the location of white particles.	195
<b>6.7</b>	Mean Dispersion in Y-coordinate for white particles (a) Packing-1 (b) Packing-2 (c) Packing-3 (d) Packing-4	197
<b>6.8</b>	Kernel density estimation for (a) Packing-1 (b) Packing-2 (c) Packing-3 (d) Packing-4	200
<b>6.9</b>	Flow map for (a) packing-1, (b) packing-2, (c) packing-3, (d) packing-4	202

## LIST OF TABLES

<b>Table No.</b>	<b>Title</b>	<b>Page No.</b>
2.1	Values of parameters for Koekemoer and Luckos's correlation (Koekemoer and Luckos, 2015)	25
2.2	Correlations for prediction of mean voidage in packed bed reactors	28
2.3	Values for Pushnov's correlation	31
2.4	Values for Benyahia and O'Neill's correlation	31
2.5	Experimental and numerical methods to measure voidage distribution	35
2.6	Correlations for the radial porosity of trickle bed reactor	41
2.7	Values of constants used in equation 2.3	45
2.8	Review of studies comparing dense and sock loading methods	46
2.9	Two-phase pressure drop models in the literature	52
2.10	Liquid saturation correlations for both low and high pressure operation	56
2.11	Mass Transfer Coefficients used in TBR	59
2.12	Studies on liquid distribution in trickle bed reactor	62
2.13	Wetting efficiency correlations for both low and high pressure	67
3.1	Summary of research contributions to the estimation of conditions to avoid wall effects	89
3.2	Experimental conditions with characteristics of the catalyst particle and column	90
3.3	Values of the parameters for first, second and third nearest neighbors	103
4.1	Characteristics of packing methods and their structural properties	128
4.2	Values of parameters of equation (4.4) for all packing methods	137
4.3	Values of mean and coefficient of variation for area distributions of all packing methods at different heights	147
6.1	Dimensions of hopper and reactor	186
6.2	Structure and flow properties of the four packing structures	192