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**Hydrodynamics of Low-Flux and High-Flux Circulating Fluidized Beds**

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Dissertation for the degree of Doctor of Science in Technology to be presented with due permission of the Department of Mechanical Engineering, Helsinki University of Technology, for public examination and debate in Auditorium K216 at Helsinki University of Technology (Espoo, Finland) on the 15<sup>th</sup> of November, 2002, at 12 o'clock noon.

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## ABSTRACT

The ongoing growth of world population and industrialization is increasing the primary energy demand, and also the need for transport fuels. The increasing carbon dioxide and other emissions are fostering the political atmosphere to demand for a more sustainable development with more efficient usage of raw materials and resources.

In order to improve some key processes in refineries, such as Fluidized Catalytic Cracking (FCC), a better understanding of high-flux flow hydrodynamics is necessary. FCC units are producing a large proportion of gasoline world-wide, and some other valuable products such as light olefins and light cycle oil. Nearly all FCC units in production utilize a riser reactor, where the solids (catalyst) circulation rate could range from 400 kg/m<sup>2</sup>s to 1,200 kg/m<sup>2</sup>s, and the superficial gas velocity from 6 m/s to 28 m/s. Therefore, it is surprising that nearly all studies of CFB pilot hydrodynamics have been carried out at low solids fluxes of less than 200 kg/m<sup>2</sup>s, whereas only a few limited but helpful studies are discussing higher solids fluxes of over 500 kg/m<sup>2</sup>s (Zhu and Bi, 1995). Such studies are regarded very useful for industrial processes and unit design, development and optimization.

In comparison to those studies carried out in the dense suspension upflow (DSU) regime (Grace et al., 1999), this Thesis will discuss high-flux operations where the axial solids holdup profile is not flat and the cross-sectional solids concentration is clearly less than 10%-vol in the upper portion of the riser. Consequently, the operation has similarities to both DSU and fast fluidization (FF) flow regimes. Since the cross-sectional solids concentration is also low (<10%-vol) in the upper portion of industrial FCC risers, another aim is to provide a detailed image of the radial solids concentration profiles and their development toward the top of a high-flux riser. Since there is confusion of how and why DSU flow regime would occupy a riser, some fundamental reasons are discussed in detail. It is shown that to realize a high-density circulating fluidized-bed operation (HDCFB), a high-flux circulating fluidized-bed (HFCFB) operation is essential but not sufficient.

For having some experience enclosed from a low-flux riser, paper I is discussing the low-flux riser hydrodynamics concerning especially the flow structure near to the column wall. Paper II and III present the solids concentration and particle velocity profiles and flow development in a long and high-flux riser. Paper IV goes on to define a novel concept of four longitudinal sections in a HFCFB riser (but not in a HDCFB riser). This concept may be a very useful fundamental aid for industrial modeling of HFCFB risers. Paper V presents some operating experience of a high-flux riser with a novel design in the solids feeding inlet. Paper VI is discussing the particle aggregation in a HFCFB riser. The collected data in papers II to VI are believed to be useful for several industrial applications since not much measured data existed under high-solids fluxes.

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## **Preface**

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Before the time in Canada I was working in industry with Fluid Catalytic Cracking development, and I still allow many thanks to I. Eilos, P. Hagelberg and Dr. J. Aittamaa for guiding me.

Finally all the people who supported me during my stay in four countries deserve many thanks.

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## Table of Contents - Part A

<b>1. INTRODUCTION .....</b>	<b>1</b>
1.1. Background .....	1
1.2. Objectives of Thesis and Definitions .....	3
1.3. Structure of Thesis.....	5
<b>2. PREVIOUS WORK AND BASIC CONCEPTS.....</b>	<b>8</b>
2.1. Powder Classification .....	8
2.2. Flow Regimes.....	11
2.2.1. Bubbling Bed Flow Regime.....	12
2.2.2. Turbulent Flow Regime .....	13
2.2.3. Fast Fluidization Flow Regime .....	13
2.2.4. Pneumatic Conveying .....	15
<b>3. EXPERIMENTAL METHODS FOR CFB RISER FLOW STUDIES.....</b>	<b>16</b>
3.1.1. Low-Flux Conditions .....	16
3.1.2. High-Flux Conditions.....	17
<b>4. LOW-FLUX CFB RISER HYDRODYNAMICS .....</b>	<b>21</b>
4.1. Solids Flow Direction in a Low-Flux CFB Riser .....	21
4.2. Axial Segregation of Particles in a Low-Flux Riser .....	23
4.3. Flow Nonhomogeneities.....	24
<b>5. HIGH-FLUX AND HIGH-DENSITY CFB RISER FLOW..</b>	<b>26</b>
5.1. Basic Concepts and Operating Conditions .....	26
5.2. Solids Concentration.....	29
5.2.1. Radial Profiles of Solids Concentration .....	29
5.2.2. Axial Solids Concentration Profiles .....	30
5.3. Particle Velocity .....	32
5.3.1. Radial Profiles of Particle Velocity.....	32
5.3.2. Axial Profiles of Particle Velocity - Particle Acceleration.....	37
5.4. Solids Backmixing.....	39

5.5.	One-Dimensional Slip Velocity .....	40
5.6.	Flow Development.....	41
5.7.	Choking and CFB System Instabilities .....	44
5.8.	Micro and Macro Flow Structures .....	47
<b>6.</b>	<b>SCALE-UP AND IMPROVEMENT OF A CFB RISER FLOW STRUCTURE .....</b>	<b>50</b>
<b>7.</b>	<b>CONCLUSIONS .....</b>	<b>53</b>
<b>8.</b>	<b>SUGGESTION FOR FURTHER WORK .....</b>	<b>55</b>
8.1.	New Applications .....	55
8.2.	Development of Current CFB Applications.....	56
<b>9.</b>	<b>REFERENCES.....</b>	<b>57</b>
<b>10.</b>	<b>ACKNOWLEDGEMENTS .....</b>	<b>65</b>

## Table of Contents - Part B

Section B consists of following papers:

- I Pärssinen J. H., Hagelberg, P., Eilos, I., Aittamaa, J., “Application of Dynamic Pressure Probe Technique to Characterize a Circulating Fluidized Bed”, Canadian Journal of Chemical Engineering, Vol. 77, pp. 299-304, April 1999.
- II Pärssinen J. H. and Zhu, J.-X., “Axial and Radial Solids Distribution in a Long and High-Flux CFB Riser”, American Journal of Chemical Engineering, 47 (10) 2197-2205 (2001).
- III Pärssinen J. H. and Zhu, J.-X., “Particle Velocity and Flow Development in a Long and High-Flux Circulating Fluidized Bed Riser”, Chemical Engineering Science, 56 (5295-5303), 2001.
- IV Pärssinen J. H., Zhu, J.-X., Yan, A.-I., Bu, J.-J., “Flow Development in a High-Flux and Long Riser”, in Fluidization X, Eds., M. Kwauk, J. Li and W-C Yang, Engineering Foundation, New York, pp. 261-268, 2001.
- V Yan, A.-J., Pärssinen J. H. and Zhu, J.-X., “Operating Experience of a High-Flux Circulating Fluidized Bed”, in Circulating Fluidized Bed Technology VII, Eds., J. R. Grace, J.-X. Zhu and H. Lasa, Canadian Society for Chemical Engineering, Ottawa, Canada, pp. 295-302, 2002.
- VI Yan, A.-J., Manyele S. V., Pärssinen J. H. and Zhu, J.-X., “The Interdependence of Micro and Macro Flow Structures under a High-Flux Flow”, in Circulating Fluidized Bed Technology VII, Eds., J. R. Grace, J.-X. Zhu and H. Lasa, Canadian Society for Chemical Engineering, Ottawa, Canada, pp. 357-364, 2002.

## *Nomenclature and Abbreviations*

C	constant
$d_p$	particle diameter, $\mu\text{m}$
g	constant ( $= 9.81 \text{ m/s}^2$ )
$G_s$	solids circulation rate, $\text{kg/m}^2\text{s}$
H	riser height, m
$\Delta P$	pressure difference, Pa
$\Delta P_{\text{cy}}$	cyclone pressure head (pressure loss), Pa
$\Delta P_{\text{db}}$	dense bed region pressure head, Pa
$\Delta P_{\text{m}}$	dynamic pressure of the flow, Pa
$\Delta P_{\text{r}}$	dilute region pressure head, Pa
$\Delta P_{\text{sp}}$	standpipe pressure head, Pa
$\Delta P_{\text{v}}$	pressure loss over solids control valve, Pa
r	radial distance from riser axis, m
R	radius of riser, m
SL	(one-dimensional) slip factor, (-)
$U_{\text{g}}$	superficial gas velocity, m/s
$U_{\text{mb}}$	minimum bubbling velocity, m/s
$U_{\text{mf}}$	minimum fluidization velocity, m/s
$U_{\text{p}}$	particle velocity, m/s
$U_{\text{slip}}$	(one-dimensional) slip velocity, m/s
z	height from the riser bottom, m

### *Greek letters:*

$\varepsilon$	voidage, (-)
$\varepsilon_{\text{bed}}$	overall bed voidage, (-)
$\varepsilon_{\text{s}}$	solids holdup, (-)
$\rho_{\text{p}}$	particle density, $\text{kg/m}^3$
$\rho_{\text{g}}$	gas density, $\text{kg/m}^3$

### *Abbreviations:*

$A_{\text{g}}$	particle aggregate
CFB	circulating fluidized bed
DSU	dense suspension upflow
FCC	fluid catalytic cracking
FF	fast fluidization
HDCFB	high-density circulating fluidized bed
HFCFB	high-flux circulating fluidized bed
LDA	laser-doppler anemometry
LFCFB	low-flux circulating fluidized bed
PSD	particle size distribution



# 1. Introduction

## 1.1. Background

The ongoing growth of world population and industrialization is increasing the primary energy demand, and also the need for transport fuels. The increasing carbon dioxide and other emissions are fostering the political atmosphere to demand for a more sustainable development with more efficient usage of raw materials and resources. The World Energy Council 17-th WEC Congress (Houston, USA, 13<sup>th</sup> to 18<sup>th</sup> September 1998) expects a 50% increase in global energy consumption during the next 20 years, suggesting that innovation and improvements are welcome and necessary, let it be in combustion or in refinery processing.

Lothar Reh (1999) collected well the current circulating fluidized-bed (CFB) applications together. The applications could be divided into three categories:

1. Non-catalytic heterogeneous high-temperature reactions, mainly with internal combustion of fuel and preferably used in power, waste-treatment and metallurgical industries.
2. Catalytic heterogeneous reactions at medium temperatures in the range of 170°C to 650°C, mainly at elevated pressures for refinery and petrochemical industries.
3. Low-temperature heterogeneous reactions, partly with catalytic component, for dry-scrubbing of waste gases from acidic and heavy metals components in power-, waste combustion and metallurgical industries.

Especially the category 2 is important because the oil industry globally treats about 3.3 million tons of crude oil per year and where a CFB riser cracking is becoming increasingly popular with a stake of more than 50% of world commercial gasoline (Yu, 1994; Reh, 1999). Besides this there are other rising catalytic reactor applications such as vapor phase catalytic oxidation of butane to maleic anhydride and production of tetrahydrofuran and butanediol by partial oxidation of n-butane (Lerou and Ng, 1996).

Table 1 summarizes some key operating conditions of two major CFB applications. In industrial FCC reactors the solids circulating rates could range from 400 kg/m<sup>2</sup>s to 1.200 kg/m<sup>2</sup>s, and the superficial gas velocity from 6 m/s to 28 m/s, increasing with the height (Zhu and Bi, 1995). That is much higher than in CFB combustion where solids circulating rate is typically less than 100 kg/m<sup>2</sup>s and the superficial gas velocity is also much lower (5 m/s to 9 m/s) at the column upper portion. An FCC unit riser is typically cylindrical compared to most combustors with a rectangular or square riser column. The circulating solids properties are also different as combustors utilize coarser sand-particles.

Table 1. Typical Operating Conditions of Two Major CFB Applications (Grace, 1990, Werther, 1994; Zhu and Bi, 1995; Berruti et al., 1995).

Parameter	CFB combustion	FCC riser
<i>Particle properties</i>		
Particle Geldart Group	B	A
Particle mean size, $\mu\text{m}$	100 – 300	40 – 80
Particle density, $\text{kg/m}^3$	1800 – 2600	1100 – 1700
<i>Key operating conditions</i>		
Superficial gas velocity, m/s	5 – 9	4 – 28 (increasing with height)
Solids circulation rate, $\text{kg/m}^2\text{s}$	10 – 100	400 – 1200
Solids concentration in the developed region, %	0.1 – 0.3	3 – 12
Average solids residence time, s	20 – 40	2 – 4
Pressure, kPa	100	$\geq 100$
Temperature in the exit, $^{\circ}\text{C}$	850 – 900	500 – 550
<i>Unit dimensions</i>		
Riser diameter, m	4 – 8 equivalent	0.7 – 1.5
Height-to-diameter ratio	< 5 – 10	> 20
Solids return column to riser diameter ratio	< 1	>> 1
Solids inventory	Low	High
Solids feeding control	Non-mechanical valve	Mechanical valve
<i>Key design concerns</i>		
Unit	Low construction and operating costs, high efficiency with low emissions	Desired yield of final products, high profitability by optimization and upgrading, safe unit operation

A large majority of CFB hydrodynamic studies have been carried out at a pilot-scale, typically at much lower temperatures and pressures compared to these two industrial reactors (Table 1). Additionally, according to Zhu and Bi (1995) and Issangya (1998) the solids circulating rate has also been typically much lower ( $<200 \text{ kg/m}^2\text{s}$ ) in a major proportion of the literature compared to industrial FCC risers and other high-flux reactors. A question can then be posed of whether that is an important factor considering that most findings from low-flux are often considered to apply to high-flux reactors. Besides addressing to that question, this Thesis is publishing a vast amount of data at high-flux operating conditions that have been exploited in previous literature in a too limited and insufficient magnitude.

## 1.2. Objectives of Thesis and Definitions

An assumption that low-flux ( $<200 \text{ kg/m}^2\text{s}$ ) flow profiles would be similar to high-flux profiles ( $\geq 200\text{-}300 \text{ kg/m}^2\text{s}$ ) and flow development holds true to some extent, but could be questioned. The first large set of evidence was published in the series of Issangya et al. (1997a,b; 1998; 1999; 2000; 2001), Issangya (1998), Liu et al. (1999) and Grace et al. (1999). This set of articles provides clear evidence that solids transport in a riser is not similar under low-flux and high-flux conditions.

In their work, it was shown that under high fluxes and high suspension densities the axial solids concentration profile was fairly flat and the solids net flow was primarily upwards at the riser wall. That was clearly different from the previous observations of fast fluidization (FF) flow regime. Thus, the operations with a solids flux higher than  $200 \text{ kg/m}^2\text{s}$  combined with a cross-sectional solids concentration of over 10% throughout the riser were defined as a Dense Suspension Upflow (DSU) flow regime by Grace et al. (1999).

In comparison to those studies carried out in the DSU regime, this Thesis will discuss high-flux operations where the axial solids holdup profile is not flat and the cross-sectional solids concentration is clearly less than 10% in the upper portion of the riser. Consequently, the operation has similarities to both DSU and FF flow regimes. Since the cross-sectional solids concentration is also low ( $<10\%$ ) in

the upper portion of industrial FCC risers, another aim is to provide a detailed image of the radial solids concentration and velocity profiles and their development towards the top of a high-flux riser.

Besides showing the flow profiles and flow development, this Thesis is also discussing the solids feeding inclined-pipe section as an important factor for achieving a stable high-flux operation in a rather large 12-m tall CFB unit (consisting of a 76-mm i.d. and 203-mm i.d. riser column and a common downcomer). The solids feeding section was modified to prevent some gas escaping upwards up to the solids control-valve or further, leading to faster and more stable CFB unit flow circulation. Only the 76-mm i.d. riser column was operated in this Thesis work.

In order to facilitate a discussion and comparison between low-flux risers this Thesis is also discussing a low-flux riser flow by showing some experimental results.

The key objectives of this Thesis could be summarized:

- Present a complete flow profile mapping regarding solids concentration and particle velocity in a 10-m tall high-flux CFB column
- Classify the riser-column into appropriate sections
- Present the trends of flow development under various high-flux operating conditions
- Discuss the findings in comparison to a recently defined flow regime; Dense Suspension Upflow (see Grace et al., 1999) and Fast Fluidization (FF) flow regime
- Discuss the requirements to establish a DSU regime in general
- Discuss the operating experience of a large CFB pilot-unit and improvement works in solids feeding design
- Present some new information of low-flux riser flow at a fast fluidization regime; describe a core-annulus structure in more details and report the flow reversals
- Discuss micro and macro flow experimental results (micro flow referring to e.g. clusters)

Based on the work of Zhu and Bi (1995) this Thesis is using the following definitions:

- Low-flux flow: solids circulation rate  $< 200 \text{ kg/m}^2\text{s}$
- High-flux flow: solids circulation rate  $\geq 200\text{-}300 \text{ kg/m}^2\text{s}$

This definition is proposed since during the last decades most studies on CFB hydrodynamics have been conducted with very low solids circulating rates not far from combustion applications operating conditions ( $G_s = 10 \text{ kg/m}^2\text{s}$  to  $200 \text{ kg/m}^2\text{s}$ ). At these low solids circulation rates a core-annulus flow structure has been often reported at FF flow regime (e.g. Rhodes, 1990; Miller and Gidaspow, 1992; Horio et al., 1988; Gajdos and Bierl, 1978; Werther, 1993 and Bai et al, 1995).

At higher solids circulation rates of  $\geq 200\text{-}300 \text{ kg/m}^2\text{s}$  it has been reported that the flow structure and hydrodynamics might become much different [see Grace et al. (1999), Issangya et al. (1997a,b; 1998; 1999; 2000; 2001), Issangya (1998), Liu et al. (1999)]. That is important because some industrial CFB applications utilize solids circulation rates from  $400 \text{ kg/m}^2\text{s}$  to  $1200 \text{ kg/m}^2\text{s}$ .

### 1.3. Structure of Thesis

This Thesis is divided into two sections, A and B. Section A is a guiding introduction to the enclosed articles. It is a section where new findings have been compared with literature, pointing out what is new information and why it is considered to be important to publish.

A short description of section A is given below:

Chapter 1 – Introduction: some background knowledge of fluidized-bed units and operation.

Chapter 2 – Previous Work and Basic Concepts: discussion of fluidization regimes and solids properties. A general understanding of flow regimes is necessary before discussing the enclosed articles.

Chapter 3 – Experimental Methods for CFB Riser Flow Studies: presents various methods and principles to study a CFB riser.

Chapter 4 – Low-Flux CFB Riser Hydrodynamics: discussion of new information found in this Thesis, at low-flux operating conditions that is more typical to e.g. CFB combustion process than for fluid catalytic cracking (FCC) reactors.

Chapter 5 – High-Flux and High-Density CFB Riser Flow: discussing new findings of flow structure in a large-scale high-flux riser, aiming at helping high-flux industrial applications design, development and optimization.

Chapter 6 – Scale-Up and Improvement of a CFB Flow Structure: discussing the limitations and problems in scale-up. Presents some industrial methods for improving the CFB riser flow structure referring to fluid catalytic cracking applications.

Chapter 7 – Conclusions

Chapter 8 – Suggestion for Further Work

Chapter 9 – References

Chapter 10 - Acknowledgements

The Thesis section B consists of six papers referred with Roman characters from I to VI. Paper I is discussing the low-flux riser hydrodynamics pointing out the solids downflow character at the riser wall. Paper II is a key paper presenting a flow mapping of particle concentration, whereas Paper III is discussing particle velocities. Paper IV is discussing the flow development in general suggesting new concepts and paper V is discussing the operating experience of a large-scale pilot CFB unit. Paper VI presents some results of micro flow structure, referring basically to particle aggregates (clusters). All papers included in this Thesis have been published or accepted for publication in the form they are presented here.

Section B consists of following papers:

- I Pärssinen J. H., Hagelberg, P., Eilos, I., Aittamaa, J., "Application of Dynamic Pressure Probe Technique to Characterize a Circulating Fluidized Bed", Canadian Journal of Chemical Engineering, Vol. 77, pp. 299-304, April 1999.
- II Pärssinen J. H. and Zhu, J.-X., "Axial and Radial Solids Distribution in a Long and High-Flux CFB Riser", American Journal of Chemical Engineering, 47 (10) 2197-2205 (2001).
- III Pärssinen J. H. and Zhu, J.-X., "Particle Velocity and Flow Development in a Long and High-Flux Circulating Fluidized Bed Riser", Chemical Engineering Science, 56 (5295-5303), 2001.
- IV Pärssinen J. H., Zhu, J.-X., Yan, A.-I., Bu, J.-J., "Flow Development in a High-Flux and Long Riser", in Fluidization X, Eds., M. Kwauk, J. Li and W-C Yang, Engineering Foundation, New York, pp. 261-268, 2001.
- V Yan, A.-J., Pärssinen J. H. and Zhu, J.-X., "Operating Experience of a High-Flux Circulating Fluidized Bed", in Circulating Fluidized Bed Technology VII, Eds., J. R. Grace, J.-X. Zhu and H. Lasa, Canadian Society for Chemical Engineering, Ottawa, Canada, pp. 295-302, 2002.
- VI Yan, A.-J., Manyele S. V., Pärssinen J. H. and Zhu, J.-X., "The Interdependence of Micro and Macro Flow Structures under a High-Flux Flow", in Circulating Fluidized Bed Technology VII, Eds., J. R. Grace, J.-X. Zhu and H. Lasa, Canadian Society for Chemical Engineering, Ottawa, Canada, pp. 357-364, 2002.

The author of this Thesis was the principal investigator for the research in papers I-IV and supported the research in papers V-VI. He wrote papers I-IV and conducted all the experimental work. He designed and built the parts improving solids inlet structure in paper V, besides designing several parts and supervising their installation to the whole unit during a period of two years (bag-house filter, pressure transducers etc.). Much of the knowledge and data obtained from

papers II-IV was used for introductory parts in papers V and VI. A-J Yan greatly supported the papers V-VI by writing the manuscripts and collecting additional data.

In addition to the papers included in this Thesis, this work has led to several other papers of which some are still waiting to be published:

Manyele S. V., Pärssinen J. H. and Zhu, J.-X., "Characterizing Particle Aggregates in a High-Density and High-Flux CFB Riser", *Chemical Engineering Journal*, accepted for publication, October 2001.

Huang, W.-X., Zhu, J.-X. and Pärssinen J. H., "Experimental Study on Solids Acceleration Length in a Long CFB Riser", to be submitted.

Manyele S. V., Zhu, J.-X., Khayat, R. E., Pärssinen J. H. and Cheng, Y., "Analysis of the Chaotic Dynamics of a High-Flux CFB Riser Using Solids Concentration Fluctuations", submitted to *Ind. Eng. Chem. Res.*, October 2001.

Yan, A. J., Pärssinen J. H. and Zhu, J.-X., "Ends Effects in a High-Flux Circulating Fluidized Bed Riser", to be submitted.

## **2. Previous Work and Basic Concepts**

### **2.1. Powder Classification**

Before going into a detailed discussion of the articles enclosed in this Thesis, it is best to consider and present first some limitations and important points for those industrials and academics who will use the information provided here. Firstly, any circulating fluidized bed (CFB) operation depends greatly on the solids properties. As this Thesis work was established with Geldart Group A particles, it limits the areas of application strongly to those industrial processes using similar solids. These applications are e.g. Fluid Catalytic Cracking



(FCC) and butane oxidation to maleic anhydride (Contractor et al., 1994; Zhu and Bi, 1995). The maleic anhydride process is having fairly similar operating conditions and riser column dimensions compared to FCC (see Zhu and Bi, 1995).

The powders could be classified in four basic groups based on Geldart (1973), see Figure 1:

Group A: particle diameter ( $d_p$ ) in most cases 30  $\mu\text{m}$  to 100  $\mu\text{m}$

- $U_{mb}$  significantly larger than  $U_{mf}$
- Large bed expansion before bubbling starts
- There is a maximum bubble size
- The bubble size can be reduced by a more wide particle size distribution (PSD) or reducing the average particle diameter
- There is a large backmixing in the emulsion phase

Group B:  $d_p$  in most cases from 100  $\mu\text{m}$  to 800  $\mu\text{m}$

- $U_{mb}$  and  $U_{mf}$  are almost identical
- No observable maximum bubble size
- Less gas backmixing in the emulsion phase
- Bed collapses immediately after (fluidization) air is shut off
- The rate at which gas is exchanged between bubble and emulsion phase is smaller

Group C:  $d_p$  in most cases less than 30  $\mu\text{m}$

- Difficult to fluidize, channeling occurs
- Interparticle forces dominant (van der Waals)
- Mechanical powder compaction
- Fluidization only possible with agitation or vibration

Group D:  $d_p$  in most cases more than 1 mm

- Mixing poor when fluidized and powders spout easily

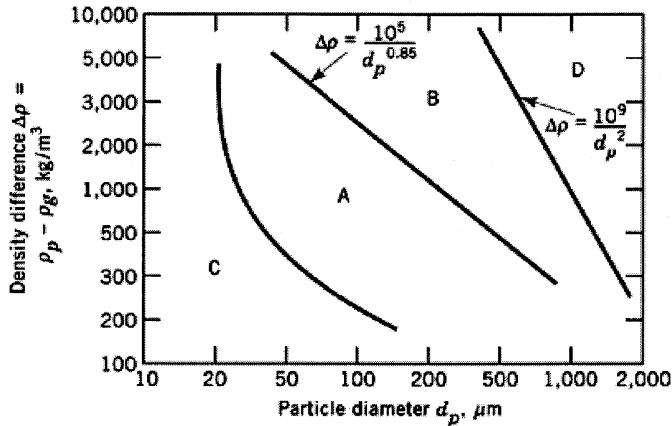


Figure 1. Geldart groups A, B, C and D (Avidan *et al.*, 2000).

The circulating solids properties have often led to operational problems in industrial CFB units. For example, in a Fisher-Tropsch (FT) synthesis demonstration unit in Brownsville (Texas) in the early 1950' the engineers were forced to abandon the use of group B particles in a 18-m tall unit due to large bubbles formation, making the unit operation unstable and fluctuating. The unit became operational later on with a Geldart Group A powder (Squires, 1994; Steynberg *et al.*, 1991).

Partly due to that, Geldart Group A powder is commonly used in FCC units to support a smooth and stable operation. Group A powder is better aeratable with a longer de-fluidization time and a smaller maximum bubble-size, leading to less fluctuating control valve differential pressures and flow rate and a more secure operation (Raterman, 1985; Geldart and Radtke, 1986). However, there might be supplementary methods for improving the solids circulating in a fluidized-bed operation. This Thesis is proposing a design for solids inlet structure that was found to be helpful for increasing the maximum solids circulating rate above 200 kg/m<sup>2</sup>s, and allowing at the same time a stable operation at high-flux conditions (Paper V). Before this change was made, the operation at high-flux conditions was not practical due to large pressure fluctuations over the solids control valve. The CFB unit riser column and the end-section of the inclined solids feeding pipe were constructed of Plexiglas to allow visual observations.

The solids used in this Thesis work were FCC catalysts obtained directly from industrial cracking units. Any large impurities such as metal pieces or small rocks were filtered away since they could accumulate on the circulating flow control valve, leading into a gradual and constant drop in the solids circulation rate. That was especially important for the Paper V experiments.

In this Thesis work the mean particle diameter and particle density are 100  $\mu\text{m}$  and 1450  $\text{kg}/\text{m}^3$  (for Paper I), and 67  $\mu\text{m}$  and 1500  $\text{kg}/\text{m}^3$  respectively (for Papers II to VI).

Besides considering catalyst physical properties for fluidization behavior in general, small size particles may be beneficial for a catalytic cracking unit economics derived from the end-products. A higher fraction of fine particles is often leading to a higher conversion in the cracking units, which might partly be related to a relatively higher surface area of catalyst available for cracking reactions. Yates and Newton (1986) and Pell and Jordan (1988) are good sources for more supporting information, suggesting that several industrial cracking units should use a high proportion of fine particles.

## 2.2. Flow Regimes

The flow regimes in industrial applications are illustrated on Figure 2. By increasing the superficial gas velocity the flow regimes develop from (a) bubbling bed to (b) turbulent regime, and finally to (c) fast fluidization for Geldart group A (or B) particles.

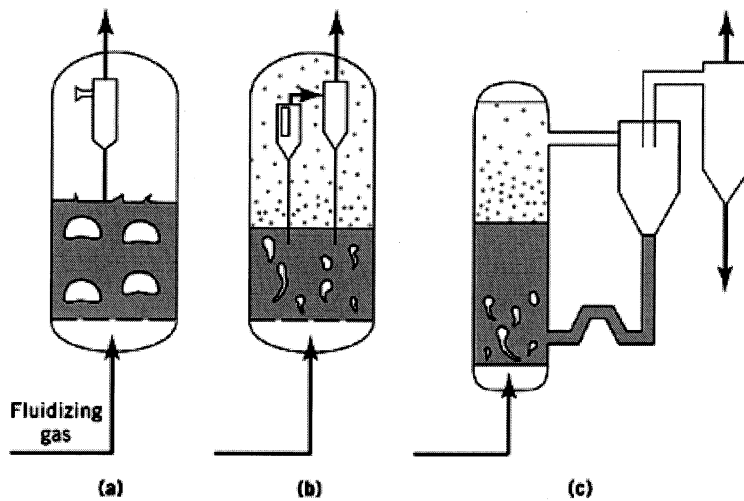


Figure 2. Flow regimes in industrial applications (a) bubbling bed regime, (b) turbulent regime and (c) fast fluidization with solids circulation (Avidan *et al.* 2000).

### 2.2.1. Bubbling Bed Flow Regime

A fixed bed expands when some gas is passing upwards via the void spaces between the particles, resulting in a greater uplifting force on the particles compared to gravity. By increasing the gas flow rate further, some excess gas is starting to go up as bubbles. This is the point where a bubbling bed regime is reached.

By increasing the gas flow rate further, bubbles start to coalesce and grow together while moving upwards. Some of the largest bubbles occupy the whole column cross-section especially with Geldart group B solids, and could be referred as slugs. This is referred as a slugging regime. In industrial reactor columns slugging (with Geldart group B solids) would normally be a very bad phenomenon since it causes unwanted vibrations and a limited gas-solid contact.

### 2.2.2. Turbulent Flow Regime

By increasing the gas flow rate further, a turbulent regime could finally be achieved. The solids suspension is dense at the column bottom, and the voidage ( $\epsilon$ ) is of the order 0.7 to 0.8 whereas the relative superficial gas velocity ( $U_g - U_{mf}$ ) is still low at 0.4 m/s to 0.6 m/s (Sun and Chen, 1989). The flow is extremely chaotic, and it is difficult to follow the constantly changing voids of gas and solids at each cross-sectional point. For FCC solids the superficial gas velocity in the column is typically below 1 m/s. The equation by King (1989) can be used to estimate the overall bed voidage for FCC solids:

$$\epsilon_{bed} = \frac{U_g + 1}{U_g + 2}$$

The axial solids concentration at turbulent fluidization regime varies significantly and fairly sharply from the dense bed region up towards the very dilute freeboard section. Brereton and Grace (1992) describe that intermittent burst of particle agglomerates become dominant over slug-like behavior at turbulent fluidization dense bed region. Illustrations of turbulent bed nature could be found from the literature (Brereton, 1987; Bi et al., 1995; Rhodes and Geldart, 1986).

### 2.2.3. Fast Fluidization Flow Regime

When the gas flow rate is increased further, more solids are entrained into the freeboard section, lowering the dense bed region solids concentration typically to 15%-vol to 20%-vol, from 20%-vol to 30%-vol that was previously presented for a turbulent regime. On the other hand, as more solids start conveying towards the column top, a higher solids concentration of <3%-vol is typically found in the riser top. Li and Kwauk (1980) were among the first ones to report a co-existence of a dense bed region and a dilute region. Yerushalmi et al. (1978) previously reported solids aggregation as clusters and strands with extensive backmixing of solids, with slip velocities much higher than the terminal velocity of the particles. Leung (1980) made a remark that pressure gradient is decreasing when the gas velocity is increased at a constant solids circulation rate (at pneumatic conveying the pressure gradient would again start to increase).

A dominant feature of fast fluidization has become that a core-annulus flow structure forms, with a solids net downflow on the wall. Several groups have measured the thickness of the downflow at low solids circulation rates of  $<130 \text{ kg/m}^2\text{s}$  (Ishii et al., 1989; Horio et al., 1988; Gajdos and Bierl, 1978; Miller and Gidaspow, 1992). These closely packed particles are referred to as clusters, streamers, strands, swarms and particle sheets depending on their shape and size (see Brereton and Grace, 1993; Horio and Kuroki, 1994). Strands and streamers could be often observed at the riser wall. There the flow is more stagnant partly due to a much lower gas velocity and forming aggregates are less disturbed allowing them to grow bigger. Closer to riser center smaller aggregates are often referred as clusters. Their size is reported to be 2mm to 15mm in most literature (see Li et al., 1991; Wei et al., 1994; Kurioki and Horio, 1994). Subbarao (1986) and Xu (1996) have proposed some correlations for calculating the diameter.

Soong et al. (1994) found that clusters seem to appear less than 15% of time at different heights of the riser. Li et al. (1996) concluded that clusters diameter most likely decreases toward the column top (with decreasing solids concentration). A higher gas velocity has likely a similar influence. Horio et al. (1992) found that by increasing the superficial gas velocity from 0.7 m/s to 1.3 m/s the cluster size decreased from 10-30mm to 3-10mm (see also Horio and Clift, 1992).

A cluster consisting of closely packed particles may have a greater "free falling velocity" compared to individual particles terminal velocity. Increasing amount of clusters may then increase the velocity difference between gas and solids, referred as a slip-velocity. That holds for gas-solids up-flow.

On the other hand, when the particles are very compactly packed together, and the gas velocity is relatively low (or particles have not been fully accelerated) the particles could actually flow against the gas flow direction. In a riser column the particles have been often reported to flow downwards, especially closer to the riser wall at low-flux conditions (FF regime). Partly for that reason a core-annulus flow concept is commonly used to distinguish it from other flow regimes.

#### 2.2.4. Pneumatic Conveying

Following the fast fluidization flow regime, a dilute pneumatic transport regime will be established by either increasing the gas flow rate or by decreasing the solids circulation rate, at a constant  $G_s$  and  $U_g$  respectively. Pneumatic transport is a flow regime where nearly all solids follow the direction of the main flow stream, let it be a vertical or horizontal transport line, with very little or no solids aggregation. As the flow is very dilute, this regime could also be called dilute phase pneumatic transport. The volumetric solids concentration is typically 0% to 2%, or in some cases up to 5% (see Issangya, 1998). With very low solids circulating rates it could, however, establish already at lower superficial gas velocities. The work of Chong and Leung (1986) is recommended here as a practical source for choking velocity correlations in vertical transport columns because they have compared six research group results. They also mention that column diameter is an important factor for the on-set of choking.

When the above classification of four flow regimes was established, there was not sufficient knowledge and experimental evidence of high-flux riser operations. For that reason all these flow regimes were summarized here to allow some comparison with the following parts of the Thesis.

### 3. Experimental Methods for CFB Riser Flow Studies

#### 3.1.1. Low-Flux Conditions

Low-flux CFB risers have often been studied by using pressure measurements along the column wall. That allows one to relate the pressure drop to the solids inventory in the respective vertical section. This is referred as apparent solids holdup, and it is shown to be somewhat higher than the actual solids holdup (Arena et al., 1996). Van Swaaij et al. (1970) found that the solids frictional pressure drop account for 20% to 40% of the total pressure drop, and at higher fluxes about 25%. That holds for narrow test columns where the influence of the wall is more important than in commercial size equipments. The basic equation for determining the apparent holdup (apparent holdup =  $1 - \varepsilon$ ):

$$\frac{\Delta P}{\Delta z} = [\rho_p(1 - \varepsilon) + \rho_g \varepsilon]$$

During work described in this Thesis a quick-closing valve technique was used to present an axial profile of solids concentration, instead of presenting pressure transducer measurement data (Paper I). The low-flux riser was separated into axial sections with rapidly closing valves, allowing one to determine the solids holdup values by weighting the solids in each section. The collected samples particle size distribution (PSD) was analyzed with a laser-diffractor to determine whether there was axial size segregation of FCC solids. Previously Dry et al. (1987) and Bodelin et al. (1994) have reported radial size segregation of solids in a CFB riser, and that might lead to axial size segregation referring to a core-annulus flow structure and solids downflow at the riser wall likely depending of solids properties.

Momentum probes and Pitot-type probes have been widely used to study gas-solids flow in CFB risers (Bader et al., 1988; Azzi et al., 1991; Yang et al., 1994; Issangya et al., 1997a). In this Thesis a low-flux riser was studied similarly with respect to flow dynamic pressure fluctuations. As variations of the flow were studied, rather than the absolute values, there was no need to calibrate the probe with respect



to constant C discussed in most literature. Constant C is a correction factor in the basic equation that could be used for obtaining dimensional results for the pressure difference:

$$\Delta P_m = C \frac{1}{2} [\rho_p (1 - \varepsilon) U_p^2]$$

For obtaining more information on constant C at low-flux conditions please refer to literature (Breugel et al., 1969; Azzi et al., 1991; Bai et al., 1996; Zhang et al., 1997). When the gas-phase dynamic pressure is ignored in the equation, the relative error could typically be less than 5% under low-flux conditions according Bai et al. (1996).

Other methods to study low-flux riser nonhomogeneities would be to use e.g. standard deviations of local particle concentration (Brereton and Grace, 1993). Particle concentration fluctuations could be measured with optic-fiber probes such as Issangya (1998). Other optical techniques for a very dilute flow would be a Laser Doppler Anemometry (LDA). In this technique light passes from a laser-source into the gas-solids flow, and then back into a collector-sensor (Berkelmann and Renz, 1991). Werther (1999) suggested that the LDA technique is not well suited for determining local solids volume concentrations, compared to a fiber-optical LDA (see Werther et al., 1996). That incorporates a probe-tip emitting laser-light inside the fluidized-bed, rather than leaving this source on the column-wall window. Werther et al. (1999) gives a good summary of all other possible gas-solid measurement techniques that are not discussed here such as video-imaging,  $\gamma$ -ray transmission tomography, capacitance-probes and suction probes.

### 3.1.2. High-Flux Conditions

Table 2 indicates several experimental techniques for a high-flux CFB pilot. The axial solids concentration profiles could be obtained from pressure transducers or quick-closing valves similarly to low-flux risers. For radial solids flux profiles a suction probe is one alternative, as the suction velocity is shown to have only a fairly small influence on the solids collection rate (Rhodes et al., 1990; Kruse and Werther, 1995). However, Schoenfelder et al. (1996) suggests quite the opposite especially for finer 50  $\mu\text{m}$  mean diameter

solids with an apparent density of  $1420 \text{ kg/m}^3$ . Fine solids passing just next to the probe tip might be aspirated into the suction probe easier than larger particles with a higher momentum. Considering other techniques, the group at the University of British Columbia has used optic-fiber probes for several publications in the end of 1990's. The high-flux pilot of this Thesis is illustrated in Figure 3.

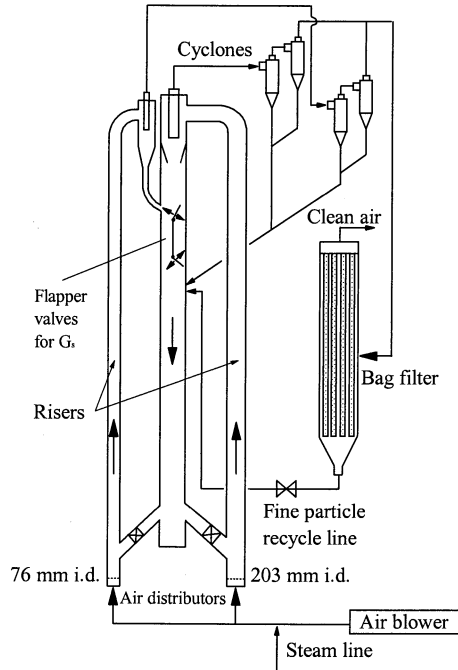


Figure 3. High-flux CFB unit (10 m tall) used in experiments at the University of Western Ontario, Canada.

The high-flux CFB system shown in Figure 3 consists of a very tall downcomer having a 350 kg inventory of FCC solids. From the downcomer solids passed through a special inclined pipe section (see Paper V) into the riser bottom. The solids were mixing with up-flowing gas distributed via a perforated plate with 2-mm openings, situated some 0.3-m below the inclined pipe entrance. The solids were transported up to the column top where a smooth exit directed the solids into the cyclone-section where gas and solids were

separated. Thereafter the solids circulation rate was measured in the downcomer with a flapper-valve system.

Table 2. Laboratory and Industrial Scale CFB Risers Experimental Techniques Where Solids Circulating Rate Exceeds 425 kg/m<sup>2</sup>s

Authors	G <sub>s</sub> (Max.) (kg/m <sup>2</sup> s)	U <sub>g</sub> (Max.) (m/s)	Particles	Technique
Arena et al. (1988)	600	7.0	Glass beads	Quick-closing valves, pressure transducers
Azzi et al. (1991)*	1080	21.0	FCC	Gammametry
Contractor et al. (1994)	590	7.8	FCC	Not reported
Liu et al. (1999); Issangya et al. (1997); Issangya (1998)	486/425	8.0	FCC	Helium gas tracing, Optic-fiber probe, Pitot-type momentum probe, pressure transducers
Karri and Knowlton (1995)	586	~13.5	FCC	Suction probe, pressure transducers
Pugsley et al. (1996)	700	8.5	Sand	Pressure transducers
This study	550	10.0	FCC	Optic-fiber probe, pressure transducers
Viitanen*	485	13.0	FCC	Radioactive tracing

\*industrial FCC unit

A five-fiber optic velocity probe was used to measure the particle velocity during this Thesis work. Such a probe consists of two light emitting fibers (B and D) and three light receiving fibers (A, C, E) arranged precisely along the same line. A particle moving by the center point between any two neighboring fibers will produce a reflective signal to a detection fiber. By counting the time difference between the two signals from A-B and B-C (or C-D and D-E), the velocity of a particle passing along the array of the five fibers can be determined.

In the above the "A-B" refers to the fact that that probe B emits the light and probe A receives the light. It is then actually only the back-coming light-signal from probe A that is used for counting the time difference. Similarly, "B-C" refers to the fact that B emits the light and probe C reflective signal is used for determining the time difference. This is to clarify the origin of the reflective light-signal in probes A and C. This probe can measure the ascending and/or descending particles (time-average) velocity, and it ignores the horizontally moving particles.

This probe does not have a fixed sampling frequency, but it samples every particle passing by and reports all qualified measurements. More details of this probe are presented elsewhere (Zhu et al., 2001). At each measurement location the sampling time was typically over 30 s, which gives a minimum of 2500 sampled particles. The particle velocity was measured on seven axial levels ( $z = 1.53, 2.73, 3.96, 5.13, 6.34, 8.74, 9.42$  m) and at 11 radial positions ( $r/R = 0.00, 0.16, 0.38, 0.50, 0.59, 0.67, 0.74, 0.81, 0.87, 0.92$  and  $0.98$ ). The horizontal line on which the measurements were done was in 90-degree angle with the solids feeding. Some flow asymmetries might exist especially at the level  $z = 9.42$  m, and their importance could be estimated by comparing levels  $z = 8.74$  m and  $z = 9.42$  m (see also publications list Chapter 1.3).

Solids concentration was measured with a reflective-type fiber-optic concentration probe. The 3.8 mm diameter probe tip consisted of approximately 8000 emitting and receiving quartz fibers, each having a diameter of 15  $\mu\text{m}$ . At each location two measurements were carried out for over a 30 s period with a sampling frequency of 1000 Hz, yielding 30 000 data points. More details of this probe could be found from literature (Zhang et al., 1998). To verify the accuracy of both probes, the local solids flux obtained by multiplying the local solids holdup and particle velocity was integrated over the cross section and compared with the average solids flux (external solids circulation rate), and it was found that the maximum deviation is less than 10-15% except from the entrance section just above the solids distributor (Zhang et al., 2001; see Paper III). The velocity probe was calibrated with a rotating disk device (Zhu et al., 2001). It had a logic circuit to select the correct data and to calculate the particle velocity. Zhou et al. (1995) achieved similarly good measurement accuracy (deviation  $<10\%$  respectively) with the same velocity probe design.

During the experiments the probes were inserted alternately into the riser, and moved horizontally into a certain radial position. The solids concentration was measured on the same axial and radial positions as the particle velocity (and at an additional elevation of  $z = 5.90$  m). The humidity level of the air was controlled between 70% and 80% to eliminate electrostatics in the system. This was carried out with a low-pressure steam-injection (and mixing) line. The fine particles were re-circulated from bag-filter back into the downcomer section for maintaining the same particle size distribution (PSD).

## 4. Low-Flux CFB Riser Hydrodynamics

### 4.1. Solids Flow Direction in a Low-Flux CFB Riser

A core-annulus flow has been reported to exist in the fast-fluidization flow regime (e.g. Rhodes, 1990; Miller and Gidaspow, 1992; Horio et al., 1988; Gajdos and Bierl, 1978; Werther, 1993 and Bai et al, 1995). According to this model, most particles seem to go downwards at the column wall, often forming particle aggregates of changing shapes such as swarms and particle sheets (Bi et al., 1993a).

Table 3 collects the results and conditions that have been used in CFB riser wall region studies. Until now, there have been mostly three factors that have been studied:

- Thickness of the downflow layer
- Particle velocity in the downflow
- Aggregates size and form

Paper I is discussing the flow behavior inside the annulus describing the percentage of time the flow was upwards and how this downflow changes by increasing the solids circulation rate.

Table 3. Laboratory CFB pilot Flow Direction at the Riser Wall at Low Solids Circulating Rates ( $< 200 \text{ kg/m}^2\text{s}$ ) with FCC solids.

Authors	Dia. (m)	$U_g$ (Max.) (m/s)	Solids ( $\mu\text{m}$ )	$z(\text{m}) / H(\text{m})$	Thickness of downflow (mm)	Velocity of downflow (m/s)
Bader et al. (1988) <sup>+</sup>	0.305	3.7	76	9.1/12.2	N/A	0.3-0.9
Gajdos and Bierl (1978)	0.076	3.2	50	1.2 to 4.0/5.8	7.2-9.9	N/A
Glicksman (1988) <sup>*1</sup>	0.10	6.5	80	N/A	N/A	1.2-2
Hartge et al. (1988) <sup>o</sup>	0.40	3.7	85	N/A	N/A	0.2-2.8
Horio et al. (1988) <sup>o</sup>	0.05	1.29	60	0.36 to 1.63/2.79	N/A	0.45-0.65
Ishii et al. (1989)	0.05	1.29	N/A	1.06/2.79	3.3-4.0	N/A
Miller and Gidaspow (1992)	0.075	3.5	75	1.9 to 5.5/6.6	1.4-11.2	N/A
Nowak et al. (1991) <sup>o</sup>	0.205	4.0	46	N/A	N/A	0.6-1.0
This study <sup>+</sup>	0.11	3.2	100	3/3.5	5.0-9.5	N/A

<sup>+</sup> Pitot-type probe    <sup>\*</sup> Video-analysis    <sup>o</sup> Optic-probe    <sup>1</sup> Sand solids    N/A : not analyzed  
z : measurement height    H : riser height

From previous literature (see Brereton and Grace, 1993; Horio and Kuroki, 1994) it cannot be stated whether the solids downward flow truly occurs at the riser wall all the time. Paper I shows that even very close to the wall some solids upflow is likely to occur, although that seems to correspond to a very low fraction of time such as 5% or less at the column top.

A more remarkable finding is that by increasing the  $G_s$  from 29 kg/m<sup>2</sup>s to 92 kg/m<sup>2</sup>s (and varying slightly  $U_g$  from 2.6 m/s to 3.2 m/s), the flow changes remarkably inside the annulus. At a lower solids circulation rate the interface between upward flow in the column center and downflow at the wall is abrupt, and it becomes more gradual by increasing the solids circulation rate.

Since previous literature has not addressed that annulus flow is not simply a continuous downward flow the Paper I proposed a simple classification. "An extensive downflow annulus" is used to refer to those radial coordinates where the upward flow is present only a small fraction of time (such as <20%) efficiently transporting solids downwards. This means that the solids net flux is clearly downwards and potentially much solids are re-circulated inside the CFB column via these radial coordinates. "A turbulent annulus" is referring to radial coordinates where the upward flow is present for a significant amount of time (20%-50%) hindering the downward solids flux. This potentially leads to a much smaller downward solids net flux in these radial coordinates and into a much smaller solids re-circulation. Paper I is discussing a circular riser column while more information of a square-column cross-section is presented by Zhou et al. (1995). In a rectangular or square column the flow structure could be more non-symmetrical.

Therefore, an extensive downflow annulus may have a significant influence on the internal re-circulation of solids depending on the radial coordinates it occupies of the whole riser column cross-section. Especially when the downward flow could be present typically up to 95% of the time much solids could be transported towards the column bottom. Several other groups have observed that this downflow takes place as large particle aggregates that are sometimes disturbed by the up-flowing solids in the core.

Besides discussing the flow structure very close to a circular column wall, it was shown in Paper I, that in a small size column there could

exist some moments of downward flow at all axial and radial positions. At the riser center, the reversals were present over 20% of the time at the column bottom, and correspondingly 8% at the riser top ( $G_s = 90 \text{ kg/m}^2\text{s}$  with  $U_g = 3.8 \text{ m/s}$ ). That is consistent with the findings of Zhou et al. (1995) who reported respectively 5% in the riser dilute region ( $G_s = 40 \text{ kg/m}^2\text{s}$  with  $U_g = 5.5 \text{ m/s}$ ). Some more information could be found from Bai et al. (1996) who measured a 3-m high column at lower solids fluxes and superficial gas velocity ( $G_s = 12.6 \text{ kg/m}^2\text{s}$  with  $U_g = 1.5 \text{ m/s}$ ). That demonstrates well that solids are not necessarily accelerated fully in the first meters and for some moments of time there could be downward going particles. Any particles going downwards increase the particle residence time in the riser column. For catalytic-cracking applications such a phenomenon is naturally not desirable due to a fast coke-formation on the catalyst in the riser column, what leads into catalyst de-activation.

## 4.2. Axial Segregation of Particles in a Low-Flux Riser

Hirschberg and Werther (1998) found preferentially larger and/or heavier particles in the downflow near the wall compared to the upflow. Dry (1987) found that radial segregation does occur in the radial direction of a low-flux CFB riser, with a preponderance of coarse particles in the annulus. Bodelin et al. (1994) obtained similar results. The previous studies have typically used heavier solids, such as a quartz sand/iron powder mixture and quartz sand (Hirschberg and Werther, 1998). This Thesis brought new information on the discussion of whether FCC solids could have size segregation in the axial direction of a CFB riser.

In Thesis Paper I a small amount of explosive was used to divide the CFB riser into four axial sections. An electric-wire launch closed rapidly quillotine-like valves and the solids trapped in each section was weighed to determine the axial solids hold-up profile. Additionally, the particle size distribution (PSD) of each sample was determined with a laser-diffractor. Some samples were measured several times to confirm the results.

Very little or no FCC solids size segregation was found by comparing the solids particle size distribution (PSD) between these four axial sections. A clear downward flow of solids was found to exist at the riser-column wall under similar operating conditions, but that did not result to axial solids size segregation. The mean particle size was apparently too small (100  $\mu\text{m}$ ) to initiate any axial solids size segregation.

### 4.3. Flow Nonhomogeneities

Paper I illustrates flow nonhomogeneities against time. The peaks and higher values of non-dimensional dynamic pressure could be assimilated to particle aggregates. It is interesting that such a simple measurement procedure already offers tools for observing the particle aggregates. Soong et al. (1994) observed clusters for a maximum of 15% of time in their riser, and results in Paper I give similar and supportive indications. In Figure 4 the measured dynamic pressures in the low-flux riser column have been reorganized into diminishing order. In Figure 4, by comparing the heights of 1.45 m (b) and 3 m (a) it can be seen that the flow reversals (when the flow direction is instantly downwards) exist at all radial and axial positions in the riser. At the bottom of the riser the reversals exist over 20% of time and at the riser top 8% respectively, when the center line of the riser is observed. The denser particle aggregates could be seen in Figure 4a and Figure 4b as higher non-dimensional dynamic pressure values (peak-values), and they present roughly less than 10% of time. That is consistent with Soong et al. (1994) showing that aggregates could appear for a maximum of 15% of time.



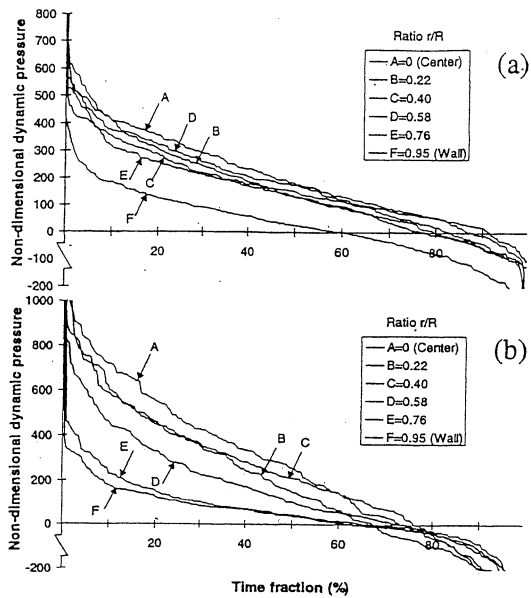


Figure 4. Flow nonhomogeneities against time (source: paper I/ Fig 6). Units are time-fraction (%) on horizontal axis and non-dimensional dynamic pressure on vertical axis: a positive value means that the flow direction is upwards in that radial coordinate in the circular column.

## 5. High-Flux and High-Density CFB Riser Flow

### 5.1. Basic Concepts and Operating Conditions

An important turning point for the understanding of high-flux CFB risers fluidization behavior was the article series of Issangya et al. (1997a,b; 1998; 1999; 2000; 2001), Issangya (1998), Liu et al. (1999). In their work, it was shown that under high fluxes and high suspension densities the axial solids concentration profile was fairly flat and the solids net flow was primarily upwards at the riser wall. That was clearly different from the previous observations of fast fluidization (FF) flow regime. Thus, the operations with a higher solids flux than  $200 \text{ kg/m}^2\text{s}$  combined with a cross-sectional solids concentration of over 10%-vol throughout the riser were defined as a Dense Suspension Upflow (DSU) flow regime (see Grace et al., 1999).

In comparison to those studies carried out in the DSU regime, this Thesis will discuss high-flux operations where the axial solids holdup profile is not flat and the cross-sectional solids concentration is clearly less than 10%-vol in the upper portion of the riser. Consequently, the operation has similarities to both DSU and FF flow regimes. Since the cross-sectional solids concentration is also low (<10%-vol) in the upper portion of industrial FCC risers, another aim is to provide a detailed image of the radial solids concentration profiles and their development towards the top of a high-flux riser. Since there is confusion as to how and why a DSU flow regime would occupy a riser, some fundamental reasons are discussed in detail in the enclosed papers. It is shown that to realize high-density circulating fluidized-bed operation (HDCFB), high-flux circulating fluidized-bed (HFCFB) operation is essential but not sufficient.

Table 4 is summarizing the operating conditions and experiments of this Thesis compared to literature. It can be seen that a more complete understanding of high-flux flow development has been obtained while papers included in this Thesis have been published.

Table 4. Laboratory and Industrial Scale CFB Risers Where Solids Circulating Rate Exceeds 425 kg/m<sup>2</sup>s.

Authors	Height (m)	Dia. (m)	Solids Feeding	Axial $\epsilon_s$	Gas mixing	Riser Flow Mapping <sup>s</sup> $\epsilon_s / U_p / A_g$
Arena et al. (1988)	6.4	0.041	Slide valve	X		No
Azzi et al. (1991)*	30.0	0.70	N/A			No
Contractor et al. (1994)	27.0	0.15	N/A	X	X	No
Liu et al. (1999); Issangya et al. (1997); Issangya (1998)	6.1	0.076	Butterfly valve	X	X	X/No/No
Karri and Knowlton (1995)	13.0	0.30	N/A	X		No
Pugsley et al. (1996)	5.0	0.05	Novel feeder	X		No
This study	10.0	0.076	Novel feeder	X		X / X / X
Viitanen* <sup>A</sup>	39.0	1.0	Slide valve			No

<sup>s</sup>CFB riser flow development has been reported completely throughout the riser,  
\*industrial FCC unit

<sup>A</sup>Viitanen study of particle residence time distribution in the reactor

Units and symbols:  $\epsilon_s$  refers to solids concentration and  $U_p$  to particle velocity,  $A_g$  refers to particle aggregates (e.g. solids concentration inside the clusters), X refers to "yes"

The operating conditions of this Thesis were varied to study the influence of solids circulating rate ( $G_s$ ) and superficial gas velocity ( $U_g$ ). Several of the studies mentioned above do not offer results for a wide range of changes in operating conditions. The most complete flow mapping so far reported by Issangya (1998) is discussing the HDCFB operation up to  $G_s$  of 425 kg/m<sup>2</sup>s with superficial gas velocities up to 8 m/s. This Thesis is presenting the HFCFB but not HDCFB operation up to higher solids circulation rates of 550 kg/m<sup>2</sup>s, while the influence of superficial gas velocity is reported up to 10 m/s. No similar study has been reported in literature presenting a complete flow mapping of a HFCFB riser, as Table 4 is suggesting. Some useful studies are mentioned in Table 4 that offer other valuable information concerning high-flux and/or high-density CFB risers. The operating conditions of this Thesis high-flux studies are shown in Table 5.

Table 5. Operating Conditions of the High-Flux Flow Riser.

Superficial Gas Vel. (m/s)	Solids Circ. Rate (kg/m <sup>2</sup> s)
5.5	300
8.0	300
8.0	400
8.0	550
10.0	300

Some key operating conditions of industrial FCC reactors could be seen in figure 5. The riser reactors superficial gas velocity is typically over 10 m/s increasing towards the riser top and the cross-sectional solid concentration could vary roughly from 2% to >10-20%.

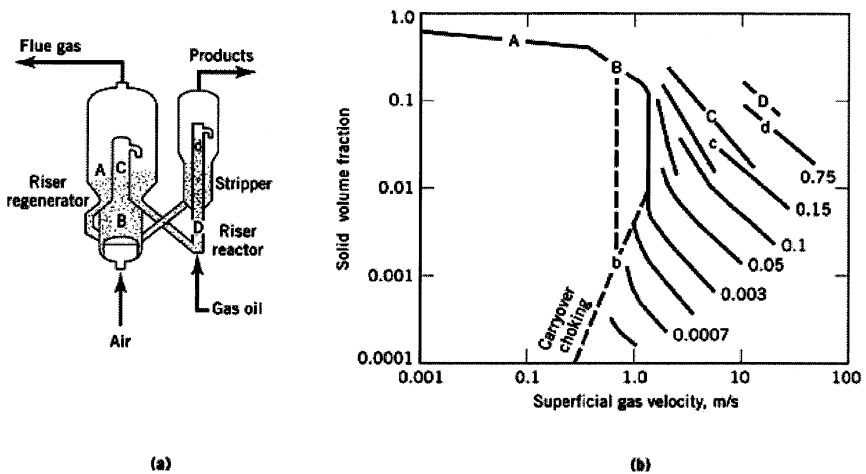


Figure 5. Industrial FCC reactor operating conditions. Small letters present the flow on the respective bed top positions. The numbers on the curves are solids superficial velocities in m/s while in modern crackers the (d) would be around 20-30 m/s (Avidan *et al.*, 2000).

## 5.2. Solids Concentration

### 5.2.1. Radial Profiles of Solids Concentration

At the riser bottom the radial solids concentration appears to have three regions under most high-flux operating conditions: a central region up to  $r/R \approx 0.5-0.6$ , where the solids holdup is low and relatively constant with a gradual increase going outwards, an intermediate region between  $r/R \approx 0.5-0.6$  and  $r/R \approx 0.8-0.9$ , where solids holdup increases significantly, and a wall region where solids concentration is extremely high (40-50%) and its increase with the radial position becomes more moderate again.

The description above is different from most low-flux risers (Bader et al., 1988; Tanner et al., 1994; Schlichthaerle and Werther, 1999). However, it is similar to the studies of Wei et al. (1998) with solids fluxes less than  $200 \text{ kg/m}^2\text{s}$ , and Yang et al. (1997) with solids fluxes less than  $130 \text{ kg/m}^2\text{s}$  and Issangya et al. (2000) in a high-density riser with solids fluxes beyond  $400 \text{ kg/m}^2\text{s}$ . The explaining factor seems to be the higher solids cross-sectional holdup in the studies the similarities were found with. It is then assumed that it is the higher solids holdups in the riser bottom ( $>0.20$ ) that determines the three-region radial structure of the solids holdups in the riser bottom section.

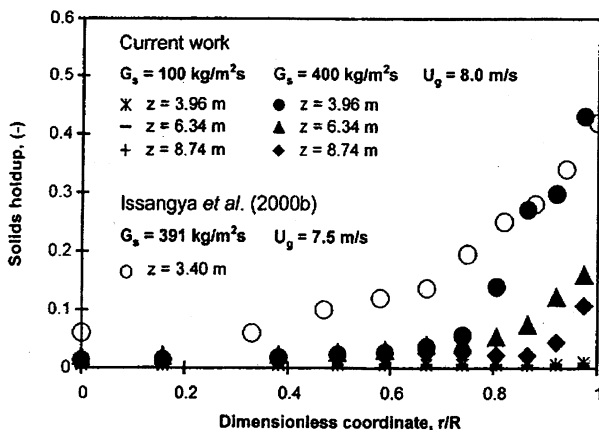


Figure 6. Radial solids holdup profiles at high ( $G_s = 400 \text{ kg/m}^2\text{s}$ ) and low ( $G_s = 100 \text{ kg/m}^2\text{s}$ ) solids fluxes (Paper II/3).

Compared to the high-density riser of Issangya et al. (2000) the Paper II profiles show similar trends up to an axial elevation of 3.4 m. At the riser top the profiles have much less in common. Paper 2 shows a wider central region and then a sharp rise in the solids holdup towards the wall compared to a much flatter profile of Issangya et al. (2000). That could be due to a much higher solids inventory and thus a higher pressure head in the Issangya group's CFB unit that could maintain a taller and denser solids suspension section in the riser. Another explaining factor is that their riser is much shorter (can avoid a dilute section). These factors are discussed in Paper II in more detail. Some similarities could be found with Karri and Knowlton (1999) regarding the HF CFB operation with a dilute section on the riser top section.

The results in Paper II are closer to the profiles that have been measured in industrial FCC risers, with similarly low solids concentrations in the riser top section, than to those of Issangya (see Martin et al., 1992; Sapre et al., 1992; Derouin et al., 1997). In industrial FCC units a much higher solids inventory would be possible to design and implement (to reach higher solids concentrations in the riser top), but also much more expensive to construct and maintain. The unit height in a refinery is often restricted partly by safety concerns (firemen access).

### 5.2.2. Axial Solids Concentration Profiles

The axial solids concentration profiles show four sections at high-flux ( $>300 \text{ kg/m}^2\text{s}$ ) conditions. These longitudinal files may be divided as follows (see Figure 7):

- Bottom dense section (I) with solids concentration of about 20% or higher
- A middle section (II) with intermediate solids concentration of about 7-8% to 15-20%
- A dilute section (III) with a typical solids concentration of 3-5%
- Top dilute section (IV) due to exit effects (which are minor in the case of a smooth exit)

For the low-flux operation ( $G_s = 100 \text{ kg/m}^2\text{s}$ ) the middle section (II) does not typically exist, so that the profile reduces to the typical S-shaped axial profile reported by Li and Kwauk (1980) and Bai et al. (1992). The only other studies that also reported such longitudinal sections are those of Issangya et al. (1997a,b, 1999) obtained under high-density (and also high-flux) conditions. However, the existence of the top dilute section cannot be seen in their study because of the shorter riser (and higher solids inventory).

For industrial designs, this finding is important and interesting because the respective heights of these four (three) sections can be found in the enclosed articles under several solids fluxes. In industrial reactors the solids flux varies widely from  $400 \text{ kg/m}^2\text{s}$  to  $1200 \text{ kg/m}^2\text{s}$ . More data is necessary to find out whether and how much the respective lengths could be influenced by the total solids inventory and the overall pressure balance in the loop of a riser plus a downcomer (standpipe).

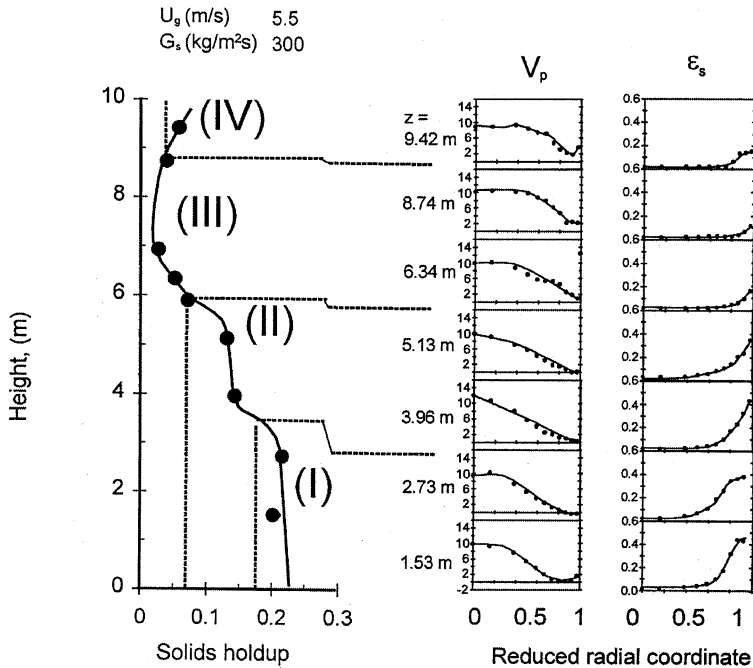


Figure 7. The four longitudinal sections and the radial solids holdup profiles in each section (Paper IV/Fig. 4).

The axial solids concentration profiles show these four longitudinal files because:

1. Radial solids concentration profile changes its shape
2. Radial particle velocity profile changes its shape

These changes are discussed and defined in Paper IV, and also here in the following paragraphs.

### 5.3. Particle Velocity

#### 5.3.1. Radial Profiles of Particle Velocity

The enclosed Paper III in this Thesis is comparing the particle velocity under low-flux ( $G_s = 100 \text{ kg/m}^2\text{s}$ ) and high-flux ( $G_s = 300 \text{ kg/m}^2\text{s}$ ) conditions under a constant  $U_g$  of 8 m/s (see Figure 8). It can be seen that under the low-flux condition the radial profiles are more uniform, and that they are less sensitive to the axial position. The latter is most likely due to a quicker flow development under low-flux conditions.

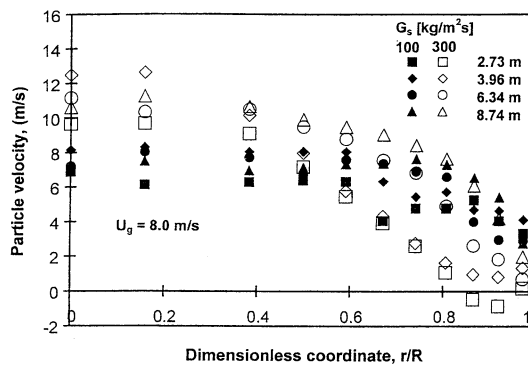


Figure 8. A comparison of the particle velocity profiles between low-flux ( $G_s = 100 \text{ kg/m}^2\text{s}$ ) and high-flux ( $G_s = 300 \text{ kg/m}^2\text{s}$ ) operating conditions at a constant  $U_g$  of 8 m/s (Paper III/2).



Unexpectedly, under higher flux ( $G_s = 300 \text{ kg/m}^2\text{s}$ ) conditions the particle velocity is higher in the column middle section (II) compared to the low flux of ( $G_s = 100 \text{ kg/m}^2\text{s}$ ) with a constant  $U_g = 8 \text{ m/s}$ . That makes sense considering that a denser concentration occupies the wall region and restricts the gas flow (see Paper II) under a higher  $G_s$  and with a constant  $U_g$ . In order to maintain a cross-sectional  $U_g$  of 8 m/s the gas velocity has to be correspondingly higher in the riser middle section. A higher gas velocity could also lead to higher particle velocities, as long as the solids concentration does not increase significantly with increasing  $G_s$  (potentially more aggregation leading to a higher local slip velocity). Knowlton (1995) indicated a similar phenomenon of a higher particle velocity at the column middle section with a higher  $G_s$  under a constant  $U_g$ . Figure 8 data could partly be explained by the fact that the actual one-dimensional gas velocity at a certain height is  $U_g/(1-\epsilon_s)$ . By increasing  $G_s$  the  $\epsilon_s$  typically increases at a certain constant height with a constant  $U_g$ .

The article series of a proposed new fluidization regime referred to as Dense Suspension Upflow [see Grace et al. (1999), Issangya et al. (1997a,b; 1998; 1999; 2000; 2001), Issangya (1998), Liu et al. (1999)] do not show particle velocity profiles as found under HDCFB operation. It would have been interesting to compare these results to Paper III, especially as the Issangya-group's column-diameter is identical to Paper III. However, as much higher solids concentrations are typically seen in the Issangya-group's column middle section, this might potentially lead to lower particle velocities in their column middle section under a certain  $G_s$  and  $U_g$  compared to the Paper III results.

The above assumption is supported by the fact that the same references show very high slip-factors for a HDCFB operation (compared to Paper II). That means that the cross-sectional mean particle velocity could be fairly low in their CFB riser, and that is not biased by the fact that they claim very little solids backmixing and flow reversals on the riser walls. It could be possible that the radial velocity profiles are flatter than in a HFCFB riser (Paper III). At the same time a higher solids suspension could hinder the individual particles from accelerating.

However, one should be extremely careful to generalize the high-flux flow hydrodynamics and preferably rely on experimental results. More studies need to be done under HDCFB operation to explain whether the suggested trends for particle velocities are correct. Any measured data would be very valuable.

Paper III and Paper IV show that four longitudinal sections exist in a sufficiently long high-flux riser. The relationship between particle velocity and solids concentration is illustrated in Figure 9.

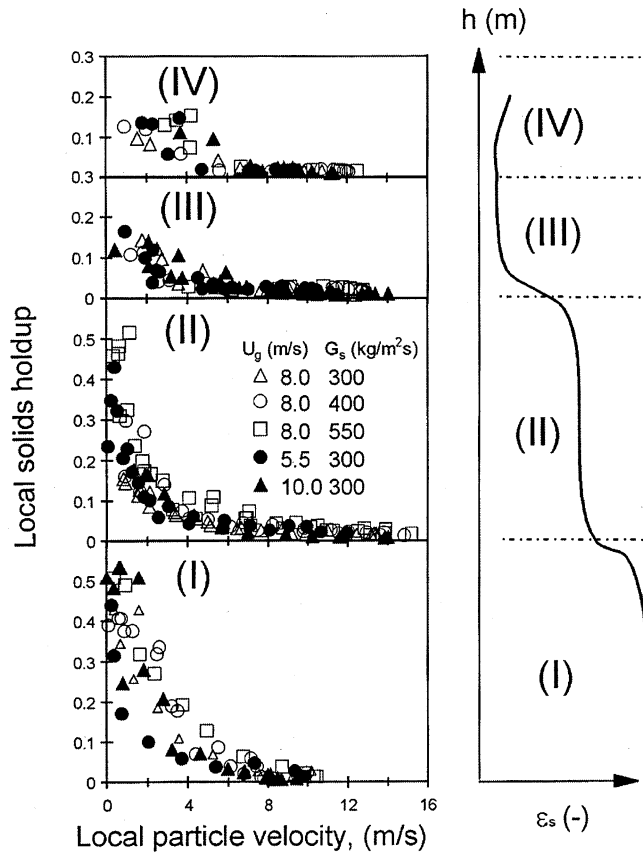


Figure 9. Relationship between (local) particle time-average velocity and solids time-average concentration in each axial section (Paper IV/6).

The four longitudinal sections are summarized in Table 6 collecting together the changes in solids concentration and particle velocity profiles.

Table 6. Radial particle velocity and solids concentration profiles in a high-flux and long CFB riser: discovery of four longitudinal sections.

Axial Sections	$\epsilon_s$	Particle Velocity Profile	Solids Concentration Profile
Bottom dense section (I)	>20%	Horizontal S-shape	Horizontal S-shape
Middle section (II)	10-25%	Linear (but not flat)	Parabolic
Top dilute section (III)	3-5%	Parabolic	Parabolic with flat center
Exit region (IV)	<10%	Similar to III, with high fluctuation	Similar to III, with high fluctuation

To make a sufficiently wide-range comparison of the Paper III and Paper IV results, it is necessary to compare the results to some low-flux downers. Downers are reactors with solids going downwards, and therefore they are often said to be ideal plug-flow reactors. Deng et al. (2002) showed particle velocity profiles in a downer under  $G_s$  of less than  $69 \text{ kg/m}^2\text{s}$  and  $U_g$  less than  $7.9 \text{ m/s}$  with FCC solids. The results show a flat radial distribution: the particle velocity is typically only about  $1 \text{ m/s}$  lower at the downer center line compared to downer wall. Based on such studies, several researchers could claim that studying up-flow (riser) pilots is not necessary or interesting as a new down-flow (downer) reactor has been discovered to be ideal. That should be discussed more in detail.

This Thesis is underlining the fact that low-flux data have very limited usefulness for high-flux flow modeling and design. The same principle holds for the downer-concept. It is necessary to get more data from high-flux downers before going into an industrial scale. Actually, it was already in 70's and 80's when Stone & Webster, Mobil and Texaco proposed a downer-reactor (Zhu and Wei, 1995; Wang et al., 1992). The reasons why this reactor-model did not become popular is certainly because there is not enough knowledge of high-flux downer-reactor hydrodynamics and design principles. It seems that most industrial refining companies have stopped their development work on downers since a rapid solids feeding is difficult (to obtain a high  $G_s$ ) and because the gas-solids mixing in the feed-

zone might not be as efficient as in a riser-reactor. A device might be needed to enhance gas-solids mixing in the downer inlet. By feeding solids into a downer-column they could easier fall in forms of “aggregates” or “strands” compared to a riser-column. A higher level of solids aggregation would lead to a lower surface area the catalyst particles could offer for cracking reactions. Roques et al. (1994) and Reh (1999) have presented similar and supporting conclusions of downer-reactors as discussed above.

However, it is likely that downers will make their way into industrial processes, while it is possible that they become popular for low-flux processes before they continue on their development path towards high-flux applications, with a time-horizon of several decades. Here is reminded that FCC processing is not the only CFB application (see a list in “Suggestions for Further Work” chapter).

As downer reactors might not become popular for high-flux CFB systems anytime soon, it is considered that it is worthwhile to present information on particle velocities and solids concentrations in a riser-column. At the same time it is proposed that a downer-reactor might be beneficial for several low-flux applications now and in the future, and more research is necessary to support and foster that.

### 5.3.2. Axial Profiles of Particle Velocity - Particle Acceleration

Solids acceleration in a long up-flow column under high-flux conditions based on measured data from several axial elevations has been addressed not at all or very little in literature. To give a starting point to this fairly new issue, Figure 10 shows indirectly the particle acceleration in three radial regions ( $r/R = 0.0 - 0.632$  riser middle;  $r/R = 0.632 - 0.894$  and  $r/R = 0.894 - 1.00$ ).

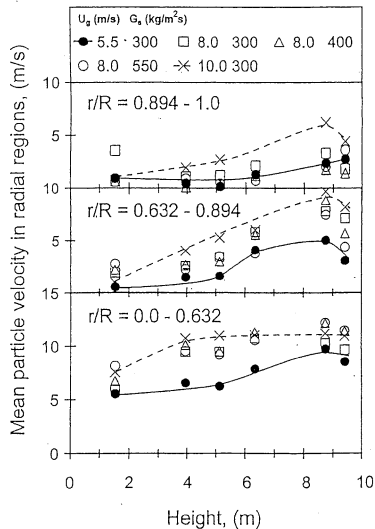


Figure 10. Particle acceleration could be derived from particle velocity measurements in various radial regions along the riser under all five high-flux operating conditions (Paper III/6).

It can be seen that in the riser middle the solids are accelerated extremely fast already at the first two meters or so. The solids continue accelerating nearly throughout the whole riser if the superficial gas velocity is low (5.5 m/s). In the radial middle section ( $r/R = 0.632 - 0.894$ ) solids acceleration at the riser top is more

significant under all operating conditions including a high  $U_g$  of 10 m/s. A clear deceleration zone can be seen below the smooth exit. In the wall section ( $r/R = 0.894 - 1.00$ ) there is very modest or nearly negligible acceleration up to a height of 5-6 meters, after which the particles very slowly start to accelerate. It can be seen that superficial gas velocity  $U_g$  can greatly influence the solids acceleration in these three radial sections. A change in solids circulation rate ( $G_s$ ) has generally speaking less influence on solids acceleration in these radial sections.

These findings are suggesting that an increase in  $G_s$  might not dramatically decrease the solids acceleration. That means that the solids acceleration in the column might remain fairly sufficient by increasing  $G_s$ , without no or no significant increase in  $U_g$ . For that reason it would be beneficial to reduce the riser column diameter in the column bottom section, leading to an increase of  $U_g$  and remarkably faster solids acceleration. Some industrial catalytic cracking reactors have already incorporated such a design.

## 5.4. Solids Backmixing

In some CFB applications, such as in fluid-catalytic cracking, the solids internal re-circulation, if any, is known to have a negative influence on the riser reactor performance. Solids internal re-circulation would result in a longer solids residence time in the reactor. In the industrial FCC risers the catalyst residence time is often designed to be around 2 seconds to ensure the desired conversion, while the actual catalytic cracking reaction may only require a fraction of a second (Avidan, 1995). Actually, this already shows that there is not enough knowledge of solids mixing patterns in the risers (Zhu and Bi, 1995), or exact confirmed knowledge of flow direction of solids at the riser wall region.

In the riser middle the gas velocity is much higher compared to the column wall and less backmixing is likely to occur. As the column wall is shown to play a major role on solids backmixing in an FF flow regime, it would be interesting to determine the particle velocities on the wall under several high-flux operating conditions. In the FF flow regime the particle velocities are generally reported to be directed downwards, but for high-flux conditions there is a lack of data.

Table 7 shows several sources indicating the flow direction in general, but none has determined the particle velocities on several axial elevations and under several operating conditions (apart from the enclosed articles in this Thesis). Even though it is shown in the enclosed articles that the time-average mean particle velocity is upwards it does not mean that there is no solids downflow under a HFCCFB operation. Some instantaneous solids downflow can be visually observed near Plexiglas-column walls under all high-flux operating conditions although solids upflow is visually observed to be more dominating. A separate article will be published on that subject by the same authors.

Table 7. Particle Flow Direction with Various Methods at a High-Flux Riser Wall.

Authors	$d_p$ ( $\mu\text{m}$ )	Dia. (m)	$G_s$ ( $\text{kg}/\text{m}^2\text{s}$ )	$U_g$ (m/s)	Method	$U_p$ at wall (m/s)
Issangya (1998)	70	0.076	200-425	4-8	Momentum probe	↑
Knowlton (1995)	76	0.20	489	11.0	Suction probe	↑
Van Swaaij et al. (1970)	N/A	0.180	200-510	4-15	NA	↑
Van Zoonen (1962)	56	0.05	625	5.5	Prandtl tube	↑↓
This study	67	0.076	300-550	5.5-10	Optic- probe	0 to 6 (up)

Remarks: Symbol (↑) refers to solids upflow and (↓) to downflow; FCC solids with  $\rho_p = 1000\text{-}1600 \text{ kg}/\text{m}^3$ , except van Zwaaij et al. (1970);  $U_p$  is measured particle velocity

## 5.5. One-Dimensional Slip Velocity

The one-dimensional slip velocity  $U_{\text{slip}}$  is generally defined to be the average gas velocity minus the average particle velocity in the column:

$$U_{\text{slip}} = U_g / (1 - \varepsilon_s) - \frac{G_s}{\varepsilon_s \rho_p}$$

In industrial design a slip factor (SL) term is commonly used, describing the average gas velocity divided by the average particle velocity:

$$SL = \frac{U_g (1 - \varepsilon_s)}{G_s / (\varepsilon_s \rho_p)}$$

In the Paper III the slip factor is found to be typically from 6 to 10 in the riser bottom and between 1.3 and 4 in the upper dilute section. Matsen (1976) reported a value of  $\sim 2$  for the dilute section of an



industrial FCC riser, while Ouyang and Potter (1993) presented a value of 2.6 at the fast fluidization regime based on pilot-scale experiments. Paper III shows that  $U_{slip}$  and thus SL depends greatly on the cross-sectional solids concentration  $\epsilon_s$  (or axial height) along with the operating conditions ( $G_s$  and  $U_g$ ). Those reporting slip factors should include some other parameters such as axial heights or operating conditions, or  $\epsilon_s$ , to allow a better  $U_{slip}$  and SL qualification for design purposes. It is clear that for industrial modeling or design of CFB reactors this parameter is one of the basic but important ones. A higher slip velocity or slip factor with the same  $U_g$  leads into a longer solids residence time in a riser-reactor. In case of FCC reactors the solid (catalyst) is de-activating rapidly and a shorter residence time could be beneficial.

Issangya et al. (1999) suggested that the solids circulation rate  $G_s$  has no influence on the slip velocity when the solids cross-sectional concentration is above 15%. Paper II (Fig. 6) does not support this entirely, showing that the influence of  $G_s$  becomes smaller but not negligible when the solids concentration goes beyond 15%. However, consistent with the observations of Issangya et al. (1999) it is shown that an increase in  $U_g$  results in an increase of slip velocities.

## 5.6. Flow Development

The article series of Issangya et al. (1997a,b; 1998; 1999; 2000; 2001), Issangya (1998) and Liu et al. (1999) are typical showing trends for high-flux and high-density flow rather than carrying out a flow mapping. Besides, there is very little change in radial solids concentration profiles when Issangya (2000) shows the levels of 0.97 m, 2.18 m and 5.23 m at  $U_g = 7$  m/s with  $G_s = 246$  kg/m<sup>2</sup>s. That is very different from the findings of this Thesis where the radial solids concentration profiles are seen to develop remarkably throughout a much longer taller riser column up to an elevation of 9.42 m, while the riser diameter is identical in both studies. More similarities could be seen if the comparison is restricted only up to a height of about 4 m or 5 m, while there still remains a surprisingly large difference in the flow development in these two riser columns. The fundamental reason is suggested to be a much larger pressure head on the two-downcomer units of the Issangya group. The dense bed region can

expand upwards in a riser if the downcomer inventory is increased. However, it would not be fair to claim that the Issangya group is operating only in a prolonged dense bed region without a dilute section. Grace et al. (1999) have clearly understood to do such a comparison, and it seems evident that it would be an oversimplification to claim DSU regime a “prolonged dense bed region” of a FF flow regime. The average cross-sectional solids concentration and particle velocities from selected articles are plotted in Figure 11a and Figure 11b showing that a clearly dilute section exists on the riser top in contrast to Grace et al. (1999). Grace et al. (1999) suggest that under similarly high solids circulation rates the whole riser would be occupied by a dense suspension upflow (10-25%-vol solids concentration).

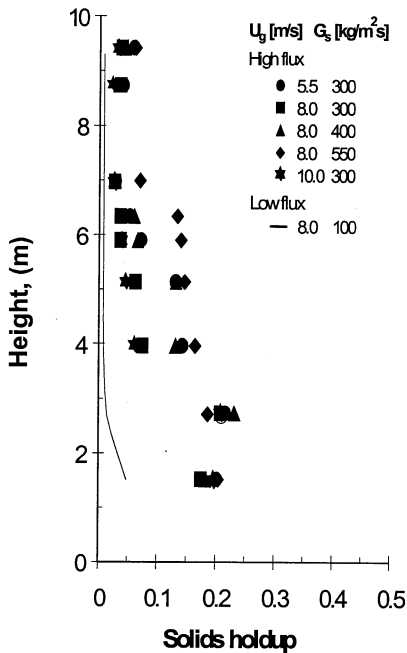


Figure 11a. Axial profile of average solid holdup for high and low solids fluxes. (Paper II/7; IV/3; V/2)

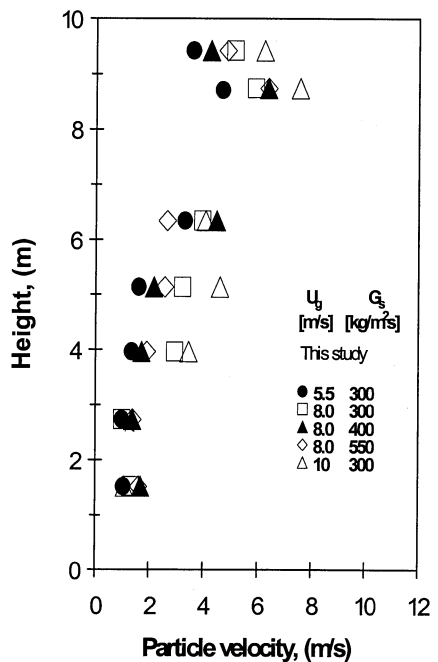


Figure 11b. Axial profile of average particle velocity for high solids fluxes. (Paper V/3; III/3a)

By increasing the solids inventory in the CFB unit the dense bed region might expand higher and higher, finally occupying the whole riser column (see Paper II; Issangya, 1998). The flow properties could then resemble more the findings of the Issangya group.

However, in industrial high-flux risers there is a rather long dilute section and therefore the data collected in this Thesis might be better applicable to current industrial units.

To come back to the findings of this Thesis, Paper II, III and IV found that radial profiles develop very differently on three radial sections ( $r/R = 0.0 - 0.632$ ,  $r/R = 0.632 - 0.894$  and  $r/R = 0.894 - 1.00$ ). For flow modeling it might be beneficial to use such an approach rather than a plug-flow model that is an oversimplification (see Fig. 12). Some engineers may have chosen to use a core-annulus flow model to characterize a high-flux riser, referring to knowledge derived from hundreds of studies carried out in CFB units at low solids fluxes. Such a scope is insufficient for high-flux flow modeling where the solids net flow direction is upwards at all radial coordinates and where the above mentioned three-radial sections work better to explain the radial and axial changes in hydrodynamics.

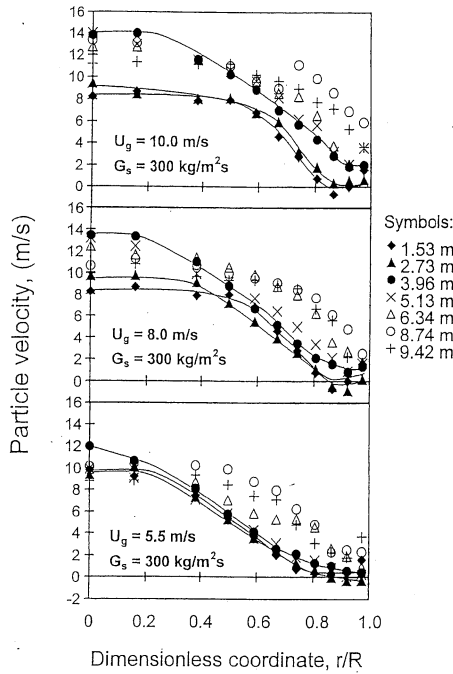


Figure 12. Development of radial profiles of local particle velocities along the riser for  $G_s = 300 \text{ kg/m}^2\text{s}$  at gas velocities of  $U_g = 5.5, 8.0$  and  $10.0 \text{ m/s}$  (Paper III/5).

## 5.7. Choking and CFB System Instabilities

A major proportion of the CFB research has been conducted at low solids circulating rates of less than  $200 \text{ kg/m}^2\text{s}$  and low gas velocities of between 2 and 8 m/s (Zhu and Bi, 1995). That is clearly related to the fact that higher solids circulation rates are difficult to achieve unless the CFB system was purposely designed to support high solids

circulation rates. A CFB pilot system consists typically of a blower, a riser, gas-solids separators, a downcomer with a certain solids inventory and a solids feeder. The system pressure balance is formed depending on these basic components.

A stable operation of a gas-solid transport line becomes impossible when the gas blower is unable to provide a sufficient pressure head (Bandrowski and Kaczmarzyk, 1981; Matsumoto et al., 1982). When the superficial gas velocity in the riser is increased, the pressure head available from a blower is normally decreasing, and this might lead into blower induced instabilities. The "blower induced instability" term was introduced by Bi et al. (1993b).

The pressure head from a blower is an important part of the pressure balance loop within the CFB system. There is a pressure loss when the solids pass through a control valve into the riser bottom, when they are conveying through the riser, and finally when they pass through the exit elbow into each of the cyclones with substantially large pressure losses (see Figure 13).

Another reason for instabilities could be, similarly, a too low solids inventory in the standpipe. Reasonably, as the gas blower and large solids inventory in the downcomer (with a proper diameter/height ratio) offer pressure heads, their sum has to be sufficient to counterbalance all pressure losses. For example, by increasing the solids inventory at a constant gas velocity (blower pressure head) and  $G_s$ , it could be possible to return from an unstable operation with some instabilities back into a stable CFB operation. Alternatively, one could obtain a higher pressure head on the downcomer side by keeping the volumetric inventory constant while switching into heavier solids, or by using a screw feeding device, (see also Bi and Zhu, 1993; Gao et al., 1991).

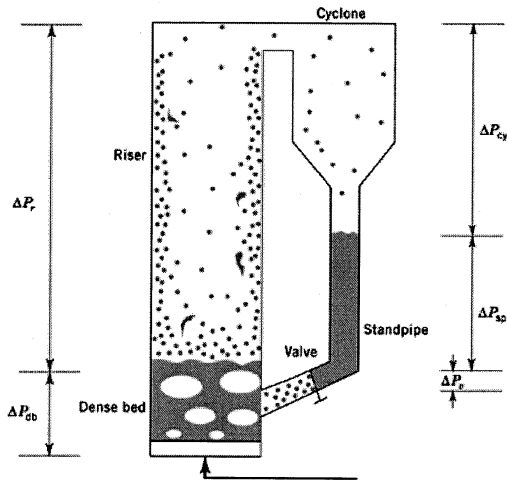
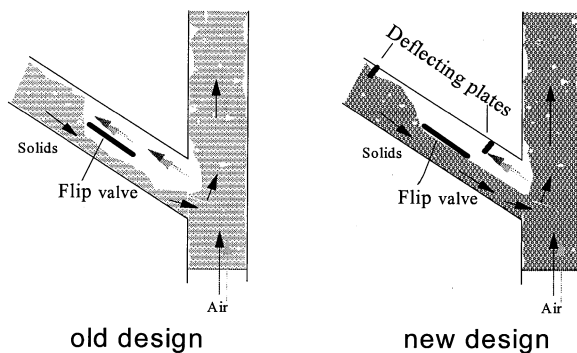


Figure 13. CFB pressure balance  $\Delta P_{sp} = \Delta P_v + \Delta P_{db} + \Delta P_r + \Delta P_{cy}$  (all pressure differences positive; Avidan et al., 2000).

The third form of choking is here called “classical choking”. When the gas flow momentum is not sufficient to carry up the solids in the riser, some solids start accumulating in the riser. This happens at a fast fluidization regime when  $U_g$  is decreased or  $G_s$  is increased leading into a slugging flow, or a bubbling or fixed bed depending of the system design and the magnitude of changes in  $U_g$  and  $G_s$ . Some systems are more sensitive than others to accept the severe pressure fluctuations resulting from slugging, and some systems can continue a relatively stable operation (Konrad, 1986).

Whereas literature suggests that a stable CFB system operation depends on the pressure loop around the unit and proper circulating powder properties, not enough attention is paid to solids feeding design. There exists no literature discussing solids feeding design similar to the one designed for the current study. Without the deflecting plates illustrated in Figure 14, some gas could escape instantaneously upwards up to the solids control valve, or further, with no objects reducing or blocking the possible gas flow. With this new design illustrated in Figure 14 the particle movement in the inclined pipe becomes much faster and steadier, and at the same time the control valve pressure difference is less fluctuating. Operation under higher flux ( $G_s = 550 \text{ kg/m}^2\text{s}$ ) is facilitated by this change.



**Figure 14.** Comparison of original and improved design of solids inlet structures of our riser (Paper V/2).

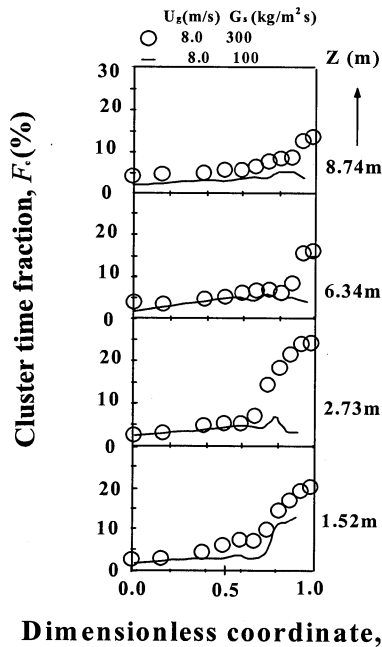
## 5.8. Micro and Macro Flow Structures

Most of literature relevant to this study discusses the macro flow structure under high-flux and high-density conditions (see Table 4 references). Macro flow is characterized by a general focus on e.g. solids holdup profiles and particle velocity profiles throughout a CFB pilot. Micro flow structure is underlining the fact that some smaller scale fluidization phenomenon takes place: existence of particle aggregates. Interestingly, this micro flow (e.g. particle aggregates) plays a major role in macro flow development, axial dispersion of solids and gas, radial distribution of solids, solids flow direction on the wall region, erosion and heat transfer on the wall, thus affecting the overall performance of a CFB riser.

The basic properties of particle aggregates are shape, frequency, size, velocity, solids holdup and time of existence. The previous literature discusses basically low-flux conditions with  $G_s$  less than  $200 \text{ kg/m}^2\text{s}$  (e.g., Horio *et al.*, 1992; Bi *et al.*, 1993a; Zethræus and Ljungdahl, 1994; Horio and Kuroki, 1994; Soong *et al.*, 1995; Horio *et al.*, 1997; Sharma *et al.*, 2000 and Lackermeier *et al.*, 2001).

Enclosed article VI compares the cluster time fraction with  $G_s$  of  $100 \text{ kg}^2/\text{s}$  to  $300 \text{ kg}^2/\text{s}$  and  $U_g$  of  $8 \text{ m/s}$  (see Figure 15). The radial profiles of cluster time fraction are less uniform under a high-flux condition

compared to a low-flux condition; and this holds for all four axial elevations towards the riser top. For that reason the data presented in article VI is useful in fostering understanding of micro-flow behavior under higher solids fluxes, as is the case in several industrial applications. The cluster time-fraction criteria were the following in paper VI: the local instantaneous solids holdup for a cluster must be greater than the time-averaged solids holdup by 1.0 to 1.4 times the standard deviation and the minimum existence time for the perturbation. The required number of consecutive samples above the critical solids holdup was taken to be three in this study. The optimal values for critical solids concentration and for the n-factor (above: 1.0 to 1.4) were obtained with a sensitivity analysis (Manyele et al., 2001).



**Figure 15.** Comparison of radial cluster time fraction profiles at high ( $G_s = 300 \text{ kg/m}^2\text{s}$ ) and Low ( $G_s = 100 \text{ kg/m}^2\text{s}$ ) solids fluxes (Paper VI/4).



Sharma *et al.* (2000) studied the solids concentration inside clusters under low-flux conditions ( $U_g$  from 4.0 m/s to 6.6 m/s with  $G_s$  of 75 kg/m<sup>2</sup>s at  $z = 4.5$  m). It suggests that clusters at the wall could be of order three times denser compared to riser middle. The article VI correspondingly shows a ratio of 7-8 under high-flux conditions at even a higher elevation of  $z = 6.4$  m. That suggests that there is a difference between these two.

A very important point article VI does not bring out is that the particle diameter could significantly influence the solids aggregation phenomena. For that reason the Sharma *et al.* (2000) is an excellent source for understanding that normal FCC catalyst particles at around 70 microns average diameter present a very different behavior compared to larger solids of 120 microns. The solids concentration in clusters is shown to vary only slightly but the cluster duration times could be longer for larger diameter solids. This makes sense because larger particles might be disturbed less by gas turbulence and other particles which might eventually break up any aggregates.

In industrial fluid catalytic crackers (FCC) the particle aggregates might have a negative influence. If oil droplets are sprayed into the bottom section of the reactor, and catalyst is flowing up in forms of large clouds of densely packed particles, some efficiency could be lost due to a hindering (shadow) effect. A better gas-solid contact would be possible if particles would not be densely packed together. For that reason the authors of this article were motivated to find a correlation between high-flux operating conditions, cluster time fraction and solids concentration inside a cluster throughout the pilot scale riser. Hopefully that serves as a base for understanding the optimization of industrial units or pre-commercial pilots micro-scale hydrodynamics.

## **6. Scale-Up and Improvement of a CFB Riser Flow Structure**

A better scale-up for CFB systems becomes possible with an increasing number of studies published in this field. A scale-up of CFB is known to be difficult and the history has shown both success and failures reported by Werther (1992) and Squires (1982). The column diameter, height, exit structure, unit pressure balance, solids feeding technique and solids Geldart Group have influence, along with parameters such as temperature and pressure. For that reason most pilot data should be discussed as beneficial information that helps understanding fundamental issues of fluidization under various flow regimes, or when doing design or optimization of industrial units, rather than being fully prepared data applicable as such for larger industrial units. Another practical problem for scale-up is that most industrial cracking unit studies are not published, or in a very limited manner, and the basis for comparing may become fairly insufficient.

A good example of the usefulness of bench-scale experiments is the evolution of the downer reactor. At the beginning the downer reactors were considered to be ideal plug flow reactors where solids were flowing downwards without backmixing. Industrial scale-up started quite fast, and designs were proposed for fluidized-bed downer cracking with an upflow riser regenerator (Murphy, 1992). It was not so evident at the beginning that the catalyst could possibly move in strands with a low interaction between the gas and catalyst, leading to somewhat under-exploitation of a full reaction surface area (Roques et al., 1994). Now the downer development continues mostly on bench-scale, mostly to address solids distribution improvements. With the steady improvements this application is already being applied in the processing industry (Zhu et al., 1995).

At the time being there is not sufficient data to determine the scale-up factors for e.g. solids concentration and velocity axial-and-radial profiles in fast fluidized-bed risers. More data is needed especially from larger pilot columns, and finally from industrial columns. As some industrial FCC riser concentration profiles have been published, it was interesting to compare them with the profiles found during this

Thesis work. The results of this Thesis work are closer to the profiles that have been measured in industrial high-flux risers (Martin et al., 1992; Sapre et al., 1992; Derouin et al., 1997) than to those of Issangya et al. (2000), whereas in their study the cross-sectional solids holdups are higher. In this Thesis the results show a wider central region with a low solids concentration and a sharper rise of concentration towards the wall. These results are also somewhat different from profiles found at low-flux conditions (see Chapter 5.2.1 Fig. 6) at the same axial CFB riser height.

For fast fluidized-beds Zhang et al. (1991) found that radial voidage profile depends solely on the cross-section-averaged voidage, irrespective of operating conditions, solids properties (Group A) and column diameters. They presented cold-pilot data for column diameters of 32 mm, 90 mm and 300 mm. As the high-flux profiles of this Thesis are rather different from the results of Issangya (1998) in a same diameter column, it could be concluded that high-flux conditions scale-up might be more challenging and uncertain. Clearly more data is necessary to conclude the work that has just begun with high solids circulating rates. It is also worthwhile to consider that Grace et al. (1999) suggested a new flow regime called high-density CFB operation (HDCFB) to underline those flow features that did not match with fast fluidization.

While the purpose of this Thesis is to explain the fundamentals of high-flux flow hydrodynamics, it is clear that one could benefit of such information while doing industrial reactor design and development. For example, Shell has improved the margin of their FCC cracking units as much as four cents (US¢) per barrel by improving their catalyst (solids) flow structure to a more 'plug-flow profile' in the riser reactor (Shell Company, 1999). Thus, a big refining company would benefit in general from such development work. All top six refining companies Conoco-Phillips, Exxon-Mobil, Valero, BP, Shell and Chevron have all a refinery crude-oil capacity of much over one million barrels per day, while a major or significant portion could go into a gasoline-mode production (FCCU) (Barrionuevo, 2002). The mentioned companies have a total market share of 59% in the United States (Barrionuevo, 2002), and understandingly they have a big influence on the economical infrastructure, as e.g. transportation costs play an important role. Thus, it is clear that the high-flux flow studies carried out in this Thesis might carry some useful information, giving the fundamentals

for engineers working with high-flux flow hydrodynamics. Considering the fluid catalytic cracking operators, another challenge is to understand how changes in operating conditions will influence the production, because right now several major refining companies are cutting their gasoline inventories. According to Barrionuevo (2002) e.g. Mobil-Exxon targets to reduce the inventories by 15%. It is clear that the production have to adjust itself to smaller and smaller inventories. That means that the operators need to understand the basic pressure-balance loop of the unit and finally how the unit can be run on the most optimal parameters. For example, the solids circulation rate ( $G_s$ ) can often be adjusted by a control-valve. During the refinery operation there is limited access to a FCC unit, mostly due to safety regulations, and therefore measurements and development are often conducted in pilot-units. Some tests that could be done on industrial units are the radioactive-tracking tests to determine catalyst residence time in the reactor, and pressure-transducer measurements on the riser column (solids concentration). The latter is fairly inexpensive, while the first understandingly is not. Some other methods such as gamma-densitometry might give useful information on radial flow structure (Werther, 1999).

To explain the impacts of this study on industrial units, the key information is simple. The suspension densities at the column wall were shown to be remarkably high under all high-flux conditions. In a good agreement with Shell Company (1999) results it is suggested that by focusing on decreasing the solids suspension density on the column wall, one might get a shorter solids residence time and a better unit performance. The radial solids concentration profiles of this Thesis work were similar to those of industrial risers (only dilute region compared), raising the question why. It is necessary to wait for more research to be published on various types and dimensions equipments before the scale-up questions could be solved.

## 7. Conclusions

This Thesis brought up new information on low-flux risers' hydrodynamics. A quick-closing valve technique that utilized small amounts of explosive to rapidly close the column into several longitudinal sections revealed that no significant FCC solids segregation by size happens in the axial direction of a CFB riser. This study also pointed out new details of a low-flux CFB riser core-annulus flow structure. Namely, the hydrodynamics of (downflow) annulus at the wall were shown to change by changing the operating conditions ( $G_s$ ). At lower solids circulation rates the interface between upward flow in the riser center and downflow at the riser wall was found to be more abrupt. The flow nonhomogeneities (due to e.g. particle aggregates) were also studied, suggesting that flow reversals exist at all riser positions under low-flux operating conditions (low  $U_g < 4$  m/s with  $G_s < 92$  kg/m<sup>2</sup>s). The fraction of time such reversals could exist was reported, and they were found to be in a good agreement with other groups' results.

The focus of this Thesis was overly on the high-flux ( $G_s \geq 300$  kg/m<sup>2</sup>s) flow in a riser column. This Thesis reported a vast amount of data of high-flux solids concentration and particle velocity profiles, showing important trends considering a newly defined flow regime (Grace et al., 1999). This Thesis is carrying more information into the discussion on this new flow regime, whereas by definition most of the current experiments were conducted slightly outside this regime. That allows also for explaining how and why a high-flux flow could exist in a high-density CFB operating range defined by Grace et al. (1999).

However, the flow structure and profiles of this Thesis are more similar to industrial units than of those other studies conducted during HDCFB operation. As a comprehensive flow mapping of solids concentration and velocity is presented for the first time for a long CFB riser at 7 or 8 axial elevations under 5 operating conditions, new classifications were presented on the flow development. The longitudinal riser sections were divided into four files, where the solids concentration and particle velocity profiles were found to be remarkably different. That concept shows that low-flux investigations have a very limited usefulness for explaining high-flux

riser hydrodynamics and flow development, or radial solids concentration and particle velocity profiles.

## 8. Suggestion for Further Work

### 8.1. New Applications

More research should be directed to those new CFB applications that are slowly coming of age, often in lack of funding and support. Reh (1999) is mentioning eight CFB applications that are in demonstration or pilot scale listed in Table 8, and who knows if several of them will reach the industrial scale. Instead of each engineer or researcher focusing on one application, cross-discipline development groups might progress much faster and solve more complicated problems, as Dr. Reh is suggesting.

Table 8. CFB Applications in a Pilot or Demonstration Scale (from Reh, 1999).

Application	Group	Development Step
Calcination of phosphate rock	High temperature non-catalytic (>400 °C)	P
Decomposition of chlorides	"	P
Combustion of coals (PCFB)	"	D, I
Gasification of coals	"	D
Hot dry-scrubbing of coal gas	"	P
Pyrohydrolysis of spent pot linings	"	P
Reduction of fine ores with coal (ELRED, DIOS, Circofer)	"	D
Treatment of steel mill residues	"	D
Butane oxidation to maleic anhydride	Heterogenous catalytic (170-650 °C)	D
Ethylen epoxidation	"	P
NO <sub>x</sub> /SO <sub>2</sub> -removal from off-gases	"	P

I = industrial                      D = Demonstration                      P = Pilot

Other interesting fields not mentioned above are fine powder research that has an increasing amount of applications in pharmaceutical industry. As the medical industry is foreseen to grow as to support the retirement of populated generations, it is obvious that there is a need for better knowledge of fine powders basic hydrodynamics and behavior, although that is not a CFB application. One other example could be the plastics recycling industry where air classification is

used to separate different density and size materials or to dry materials in a fluid-bed (not CFB applications). As major legislation changes have now occurred in the European Union such fields are seen to grow and currently there is a lack of research. Much of the knowledge obtained with CFB research and development projects might be applied to other fields of solids transport or hydrodynamics in general. Nowadays there are not sufficient efforts to cross-fertilize the knowledge and launch projects with a wider range of sponsorships.

## 8.2. Development of Current CFB Applications

For further work related to FCC risers it would be necessary to compile a summary of industrial CFB operating problems. Nowadays an organization called FCC network where general industrial unit operating problems are considered and discussed, but that remains basically unpublished.

Most industrial problems have their roots in very basic concepts of hydrodynamics and engineering. There is basically no literature discussing the industrial CFB-units' most typical operating problems and solutions that would certainly interest most readers.

Other areas of development are the solids flow structure in catalytic cracking reactors, often beneficial from economical points of view, and to moderate the consumption of primary energy by a better process.

A CFB combustion or gasification process is also an important subject, being among the two most important industrial applications of CFB in general. The developing countries install at increasing number new power facilities based often on fossil energy resources. The hydrodynamics during the combustion process is a first key factor leading potentially to lower emissions that partly could be captured using various technologies. It is obvious that much more knowledge will be discovered by several universities and industry in such topics not forgetting that there are legal, political, economical and technological issues and constraints to consider.



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