# ABATEMENT OF SULFUR DIOXIDE FROM INDUSTRIAL FLUE GASES IN A COUNTER-CURRENT MULTI-STAGE FLUIDIZED BED REACTOR

Thesis submitted to Indian Institute of Technology, Kharagpur For the award of the degree

of

**Doctor of Philosophy** 

under the guidance of

Chittaranjan M

Prof. B. C. Meikap



# DEPARTMENT OF CHEMICAL ENGINEERING INDIAN INSTITUTE OF TECHNOLOGY KHARAGPUR NOVEMBER 2008

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# List of Symbols

A	cross sectional area of the column $(m^2)$
$A_0$	pre-exponential factor (m <sup>3</sup> /mol·s)
Ar	Archimedes number (-)
$C_{I_1}C_2$	constants (-)
$C_b$	sulfur dioxide concentration of bubbles $(mol/m^3)$
$C_{bi}$	sulfur dioxide concentration of bubbles in $i^{th}$ stage (mol/m <sup>3</sup> )
Ci	sulfur dioxide concentration of gas leaving the $i^{th}$ stage (mol/m <sup>3</sup> )
$C_{i+1}$	sulfur dioxide concentration of gas entering the i <sup>th</sup> stage (mol/m <sup>3</sup> )
$C_{in}$	sulfur dioxide concentration of gas at the inlet (mol/m <sup>3</sup> or ppm)
$C_{out}$	sulfur dioxide concentration of exit gas (mol/m <sup>3</sup> or ppm)
$C_n$	sulfur dioxide concentration of emulsion phase $(mol/m^3)$
$C_{pi}$	sulfur dioxide concentration of emulsion phase in $i^{th}$ stage (mol/m <sup>3</sup> )
$\frac{-p_i}{C}$	average sulfur dioxide concentration of emulsion phase in i <sup>th</sup> stage
0 pl	$(mol/m^3)$
$C_{R}$	calcium hydroxide concentration of emulsion phase $(mol/m^3)$
$C_{P_i}$	calcium hydroxide concentration of emulsion phase in i <sup>th</sup> stage
- 10	$(mol/m^3)$
$\overline{C}$	average calcium hydroxide concentration of emulsion phase in i <sup>th</sup>
U <sub>Ri</sub>	stage (mol/m <sup>3</sup> )
$C_{R0}$	calcium hydroxide concentration of emulsion phase entering
- 110	$(mol/m^3)$
$C_{SO2}$ in	inlet concentration of $SO_2$ (ppm)
$C_{SO2}$ out	outlet concentration of $SO_2$ (ppm)
$C_d$	coefficient of discharge(-)
Ď	diameter of reactor (m)
$D_{h(z)}$	bubble diameter at height $z(m)$
$D_{ha}$	initial bubble diameter (m)
$\frac{1}{D}$	average bubble diameter (m)
$D_b$	diffusivity of SO <sub>2</sub> in gas phase $(m^2/s)$
$D_{g}$	equivalent channel diameter (m)
d	orifice diameter of the distributor (m)
$d_{r}$	particle diameter (m)
ď	particle diameter of hinary mixture (m)
$rac{m}{p}$	estimation of an atting (1-1/mal)
	activation energy of reaction (KJ/mol)
Г Б	adhasian faras (N)
r <sub>H</sub> f	adhesion force (N)
J G	Inclion factor (-) $(1 - 1)^{-1}$
$G_{mf}$	superficial mass velocity of air at minimum fluidization (kg/m $\cdot$ s)
Ga	superficial mass velocity of air $(kg/m^-s)$
Gs	superficial mass velocity of solids (kg/m <sup>-</sup> s)
g	acceleration due to gravity (m/s <sup>2</sup> )
H	height of the bed (m)
Hmf	height of the bed at minimum fluidization (m)
$n_W$	weir height (m)
n <sub>S</sub>	solids height in downcomer (m)
K	number of reaction units (-)

$K_G$	mass transfer coefficient per unit surface area of bubble $(m/s \cdot m^2)$
$K_l$	constant (-)
$k_I$	reaction rate constant (m <sup>2</sup> /mol·s)
$m_1, m_2$	constant (-)
N	number of orffices per unit area of distributor (m <sup>2</sup> )
$N_b$	number of bubbles per unit bed volume (m <sup>3</sup> )
n	number of channels (-)
p	pitch (m)
$\underline{Q}$	inter-phase mass transfer flux (m <sup>3</sup> /s)
R	ideal universal gas constant $(J/mol \cdot K)$
r	rate of SO <sub>2</sub> removal (mol/m <sup>3</sup> ·s)
r	average rate of SO <sub>2</sub> removal (mol/m <sup>3</sup> ·s)
$r_i$	average rate of SO <sub>2</sub> removal in i <sup>th</sup> stage (mol/m <sup>3</sup> ·s)
<i>Re<sub>mf</sub></i>	Reynolds number at minimum fluidization (-)
$Re_G$	Reynolds number of gas (-)
t	thickness of distributor (m)
t	mean residence time of solids (s)
Т	absolute temperature (K)
$T_i$	inlet temperature of gas (air) (K)
и	superficial velocity (m/s)
$u_b$	bubble rise velocity (m/s)
<b>u</b> op	superficial operating gas velocity (m/s)
$u_{mf}$	minimum fluidization velocity (m/s)
$u_{mb}$	minimum bubbling fluidization velocity (m/s)
$u_o$	empty tower gas velocity (m/s)
$u_{slip}$	gas-solid relative velocity (m/s)
<i>u<sub>st</sub></i>	gas velocity in downcomer (m/s)
ut	terminal settling velocity of solids (m/s)
ug	superficial gas velocity (m/s)
us T/	superficial solid velocity $(m/s)$
V b MZ	volume of bubble (m <sup>-</sup> )
V V	solids hold-up (kg)
Л 7	number of transfer units (-)
4 Guaak	bed height (m)
Ureek lattang	
lellers	
ß	fraction of gas associated with hubble phase (-)
۲ ۶	dimensionless constant (-)
7	$\frac{1}{1} = \frac{1}{1} + \frac{1}{1} + \frac{1}{2} + \frac{3}{2}$
Ps	particle density (kg/m) $f_{\rm res}$
$\rho_s$	particle density of binary mixture (kg/m)
$\rho_{a/\rho_{g/}}\rho_{f}$	density of air (kg/m <sup>-</sup> )
E <sub>mf</sub>	porosity of bed at minimum fluidization
e Ø	porosity/voidage of bed
$\omega_{\rm s}$	sphericity of solids
$ ho_L$	density of manometric liquid (kg/m <sup>3</sup> )

- enthalpy of manometric liquid (kg/m<sup>°</sup>) enthalpy of reaction (kJj/mol) difference in manometric liquid head (m) differential pressure (N/m<sup>2</sup>) ∆h
- $\Delta P$

$\Delta P_b / \Delta P_s$	differential pressure due to bed $(N/m^2)$
$\Delta P_c$	differential pressure due to constriction in downcomer $(N/m^2)$
$\Delta P_p / \Delta P_d$	differential pressure due to distributor (N/m <sup>2</sup> )
$\Delta P_{dc}$	differential pressure due to solids in downcomer $(N/m^2)$
$\Delta P_T$	total pressure drop across a stage (N/m <sup>2</sup> )
$\Delta P_w$	pressure drop due to wall $(N/m^2)$
$\Delta z$	differential height (m)
η	removal efficiency
$\eta_1$	removal efficiency of SO <sub>2</sub> at first stage
$\eta_2$	removal efficiency of SO <sub>2</sub> at second stage
$\eta_3$	removal efficiency of SO <sub>2</sub> at third stage
$\eta_n$	removal efficiency of SO <sub>2</sub> at n <sup>th</sup> stage
η 502	SO <sub>2</sub> removal efficiency
Nth (SO2)	theoretical removal efficiency of SO <sub>2</sub>
γ	kinematic viscosity (m <sup>2</sup> /s)
$\mu_g/\mu_a$	gas viscosity (kg/m·s)
$\overline{\tau_i}$	mean residence time of solids in each stage (s)
$\theta_e$	dimensionless experimental mean holding time(-)
$\theta p$	dimensionless predicted mean holding time (-)

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### Abstract

A multi-stage fluidized bed reactor (MFBR) with downcomer was designed and fabricated to study the stable operating range in respect of gas and solids flow rate, mechanism of gas-solid contacting and measure the overall pressure drop, holdup under different flow rates. Experiments were conducted on reactor to measure the gas pressure drop at gas flow rates ranging from  $31.2 \times 10^{-2}$  to  $56.4 \times 10^{-2}$  kg/m<sup>2</sup> s and solids flow rate ranging from 35.4 x  $10^{-3}$  to 141.5 x  $10^{-3}$  kg/m<sup>2</sup>·s under various weir heights. The solids taken for investigation was hydrated lime and sand. The regime in each bed was bubbling regime whereas solids regime in each downcomer was like moving bed. The pressure drop due to lime particles decreased with increase in the gas flow rate and increased with increase in solids flow rate. The minimum pressure drops occurred in each stage at high gas flow rate corresponding to lowest solids flow rate  $(35.4 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s})$  is 57.1, 103.3 and 143.5 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. The maximum pressure drops occurred in each stage at lowest gas flow rate  $(31.2 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s})$  corresponding to maximum solid flow rate (141.5 x  $10^{-3}$  kg/m<sup>2</sup>·s) are 98.4, 139.6 and 185.1 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. Similar trend also observed in case of sand particles. At ambient condition, the percentage removal efficiency of sulfur dioxide ( $\eta_{SO2}$ ) by hydrated lime particles was maximum at top stage and minimum at bottom stage. It was observed that lowering the superficial gas velocity at a particular solid velocity had higher sulfur dioxide removal efficiency of 65% at 0.07 m weir height and 62% at 0.03 m weir height, which was due to higher gas residence time. A mathematical model has been proposed and EGPF model agreed well with experimental data.

### Keywords:

Hydrodynamics; multi-stage fluidized bed; air pollution control; sulfur dioxide removal; pressure drop; hold-up; minimum fluidization velocity; residence time; flue gas

# **CHAPTER 1**

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### **INTRODUCTION AND LITERATURE SURVEY**

### Introduction and literature survey

#### **1.0 Introduction**

Air pollution is caused by the presence of one or more pollutants in the atmosphere. These pollutants are primarily divided into two categories viz. gases (sulfur dioxide, carbon monoxide, oxides of nitrogen, hydrogen sulfide, ammonia, ozone, hydrocarbons etc.) and particulates (dust, mist, smokes, fly-ash etc.). In the recent years, "air toxics" or hazardous air pollutants have gained special importance due to their potentially obnoxious effects not only on human health but also on the overall ecosystem. Additional atmospheric effects, which have become of significant concern, include photochemical smog, acid rain and global warming. Within recent years, in central Europe and several Scandinavian countries, along with Canada and north-eastern United States, attention has been directed to potential environmental consequences of acid precipitation, which are thought to be the main cause of forest destruction in these areas. Causative agents in acid rain formation are typically associated with sulfur dioxide emissions and possibly, nitrogen oxide emissions. From a worldwide perspective, sulfur dioxide  $(SO_2)$  emissions are the predominant precursor of acid rain formation, which is now an international or trans-boundary problem. Gaseous pollutants like sulfur dioxide that are once emitted into the atmosphere have no boundaries and become uncontrollable and caused acid deposition in the country where it has not been necessarily emitted. Several toxicological/epidemiological investigations during the last few decades have shown that the effect of this gas individually very severe. Consequently, in recent years, a number of studies have been undertaken for developing a better understanding of the effects of sulfur dioxide on humans, vegetations, animals and materials. The farreaching consequences and magnitude of the problem have motivated to all stakeholders such as public and private sector industries, government agencies and environmental engineers to work out on reliable background information, enforceable regulations and develop viable control options for sulfur dioxide emission. Furthermore, considering the importance and seriousness of the problem, increasing attention is also being paid to develop suitable processes and equipments for the

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control of sulfur dioxide emission, which can effectively meet the demands of technology and increasingly stringent environmental laws and regulations as prescribed by regulatory agencies.

### 1.1 Sources and effects of sulfur dioxide gas

Environmental degradation by sulfur compounds remains a major problem in developing countries like India. The emission of these compounds always occur in varying quantities and composition from various industries, e.g. pulverized coal fired thermal power plants; roasters and smelters of copper, zinc and lead; petroleum refineries; fluidized bed catalytic cracking units; sulfuric acid plants etc. Copper smelters and petroleum refineries are perhaps two major non-combustion sources of sulfur dioxide. Coal or oil fired power stations account for more than 50% of the sulfur dioxide emissions. Table 1.1 presents detail sources of few sulfur dioxide emitting industries, current Indian emission standards and existing control technologies.

Major type of industries	Load / mass based standard of SO <sub>2</sub> (Kg/ T of product)	Concentration based standard of SO <sub>2</sub> (mg/ Nm <sup>3</sup> )	Typical SO <sub>2</sub> emission from the industries	Current Treatment practice
	Old New	Old New		
Sulfuric acid plants i) C* < 300TPD** ii) C>300TPD	2.5 2.0 2.0 1.5	1370 1250 1250 950	1500-2000 mg/m3 (600-800 ppm)	Wet method (absorption by caustic in venturi scrubber at only offset condition)
Copper, zinc and lead smelters i) C< 300TPD ii) C>300TPD	2.52.02.01.5	1370 1250 1250 950	1500-2000 mg/m3 (600-800 ppm)	Wet method (absorption by caustic in venturi scrubber at only offset condition)
Coal fired thermal power plants	No standard standard is in p of Environm Government of	(Stipulation of process by Ministry ent and Forest, `India)	1100-1300 mg/m3 (450-500 ppm)	Dispersion by high Stack

Table 1.1. Sources of sulfur dioxide and its control techniques

\*C - Production capacity \*\*TPD- Tons per day

In addition to above industries, the sulfur dioxide gas is also being emitted from other industries where raw material inherently contains sulfur compounds or sulfur itself is used as basic ingredient in the process.

The effect of sulfur dioxide on overall ecosystem is enormous. It is one of the common and poisonous gases among the gaseous pollutants that are emitted from various industrial processes. It is a colorless, non-flammable gas having characteristic, irritating pungent odor and a vapor pressure of 3.2 atmosphere and 2.6 times heavier than air and 10% soluble in water by weight. It is present in low concentration in the community air as well as in higher concentration in some work places. Inhaled SO<sub>2</sub> can easily be hydrated in the respiratory tract to produce sulfurous acid, which subsequently dissociates to form bisulfate and sulfite derivatives. These derivatives can be absorbed into the blood or other body fluids. Few researchers have studied the effect of this gas on animals. It has been observed that sulfite contents increase in lungs and other organs from SO<sub>2</sub> exposure mice [Meng et al., 2005]. Epidemiological evidence has linked SO2 exposure with respiratory tract disease and lung cancer [Nyberg et al., 2000]. Increased frequencies of chromosome aberrations (CA), sisterchromatid exchange (SCE) and micronuclei (MN) have been detected in peripheral blood lymphocytes from workers chronically exposed to SO<sub>2</sub> in factories [Meng and Zhang, 2002]. Inhalation exposure to SO2 also increases the frequencies of CA and MN in mouse bone marrow cells in vivo and bisulfite and sulfite induce CA, SCE and MN in cultured human lymphocytes [Meng and Zhang, 2002]. SO2 inhalation may cause changes of oxidative stress and anti-oxidation status in various organs of mice, brains of rats and guinea pigs and the erythrocytes of rats [Haider et al., 1981].

At different concentration, its effect on human health is different. It is considered immediately dangerous to life and health (IDLH) at 100 ppm. In contrast to other irritant gases, such as nitrogen dioxide and ozone, the irritant effects of sulfur dioxide are instantaneous. Because of its high water solubility, the nausea-pharynx and upper respiratory mucous are most vulnerable to the effects of sulfur dioxide. At the level of 0.6 ppm to 1.0 ppm, SO<sub>2</sub> irritates eyes, nose and throat [Summer and Haponik, 1981]. At the relatively low concentration, the majority of SO<sub>2</sub> gas is deposited in the nose and oropharynx. However, at high level of exposures, the capacity of the oropharyngeal mucous to absorb SO<sub>2</sub> gas may be exceeded and damage to the trachea, larynx, bronchi and alveoli may result. At very high levels of exposure, upper airway obstruction, secondary edema and even death by asphyxiation may occur. In addition to these effects,  $SO_2$  may contribute to the development of pulmonary edema in some individuals. Clinically, these people complain of chest discomfort; dyspnea; marked burning of eyes and a prominent cough. A physical examination may reveal conjunctivitis, corneal burns, an erythematous pharynx and sign of pulmonary edema. The effect of chronic poisoning as a result of long term exposure to  $SO_2$  remains unclear. However, a high level of  $SO_2$  seems to result in poorer health and shorter life spans. The frequency and severity of bronchitis and respiratory disease are strongly correlated with  $SO_2$  concentrations. In general, the health consequences of  $SO_2$  are related to the highly irritant effects of sulfate aerosols (Particles), such as sulfuric acid, which are produced from  $SO_2$ . The concentration of absorbed  $SO_2$  is higher in aerosols and particulates. The effects of  $SO_2$  on human health life, animals and vegetation and building materials have also been reported by other researchers [Galea, 1964; Brandt and Heck, 1968; Stokinger and Coffin, 1968; Goldsmith, 1968, Key, 1977].

National Institute for Occupational Safety and Health [NIOSH, 1994] and the American Conference of Governmental Industrial Hygienist [ACGIH, 1996] have set the recommended exposure limit (REL) and threshold limit value (TLV) for sulfur dioxide at 2 ppm and a short-term exposure limit of 5 ppm. The Occupational Safety and Health Administrator (OSHA) has set the permissible exposure limit (PEL) at 5 ppm. National Ambient Air Quality Standard (NAAQS) of SO<sub>2</sub> in India is given in Table 1.2.

Pollutant	Time WeightedAverage	Concentration in ambient air		
		Industrial area	Residential, Rural and Other area	Sensitive area
Sulfur dioxide (SO <sub>2</sub> )	Annual Average*	80 µg/m <sup>3</sup>	60 μg/m <sup>3</sup>	15 μg/m <sup>3</sup>
	24 hours**	120 $\mu$ g/m <sup>3</sup>	80 μg/m <sup>3</sup>	$30 \ \mu g/m^3$

 Table 1.2. National ambient air quality standards (India)

\* Annual arithmetic mean of minimum 104 measurements in a year taken twice a week 24 hourly at uniform interval.

\*\* 24 hourly/ 8hourly values should be met 98% of the time in a year. However,
2% of the time, it may exceed but not on two consecutive days.

### Effects of sulfur dioxide at Paradeep

Paradeep is one of the important ports of Orissa located in the eastern coast of India. Two phosphatic fertilizer plants namely M/s Indian Farmers Fertilizer Cooperative Ltd. (IFFCO) and M/s Paradeep Phosphate Limited (PPL) have been operating in the area having captive sulfuric acid plants (SAP). The SAP of M/S IFFCO is one of the largest sulfuric acid plants in the world. Since both the plants have double conversion and double absorption (DCDA) technology for production of sulfuric acid, the DCDA technology takes care of existing emission standards at stable operating conditions. These plants operate alkali scrubbers during start-up and shut down conditions only (unstable conditions) due to higher emission during these periods. But when both the plants operate at full capacity, these plants emit around 20-22 ton of sulfur dioxide to the environment even after meeting the prescribed emission standard, out of which 35-40 ton sulfuric acid (98% conc.) could be produced if not allowed to emit to environment. At slightly unstable meteorological conditions, the emission from both the plants causes lot of damage to the environment. Further many steel plants like M/s Pohang Iron and Steel Company (POSCO) and M/s Essar Steel have started the construction of their plants at the same location. So the situation is expected to worsen further, when all the existing and new plants shall be in operation. Since the major emitter of sulfur dioxide are the fertilizer plants, these plants have to reduce the emission further and install suitable low cost SO<sub>2</sub> emission control technology since the existing DCDA technology can not reduce the emission further below from the present standard i.e.  $2 \text{ kg of } SO_2 / \text{ ton of acid}$ produced. Sometimes at even stable meteorological condition, SO<sub>2</sub> plume touches the ground surface and damages vegetation over an area up to 15 km (aerial distance) from the plant. Therefore it is very essential to reduce the emission discharge rate to a much lower level by adopting appropriate pollution control devices.

#### 1.2 Control methods for abatement of sulfur dioxide emission

To control  $SO_2$  emission, one will have to rely in the near future, mainly on waste gas clean-up processes, since development of sulfur-free emission technology will require major improvement in technology. Conventional technique of using tall stacks with no treatment of the stack gas at power stations may improve air quality locally, by dispersing the air pollutants, but is likely to aggravate the conditions in remote areas. Other industrial installations such as sulfuric acid plants having double conversion and double absorption (DCDA) technology generally use alkali scrubber during unstable condition. In fact, there is increasing concern over the total amount of  $SO_2$  emitted and the resulting increase in ambient concentration over wide areas. Stack gas cleaning to remove  $SO_2$  is therefore, likely to become a major technology over the next few decades for the reduction of  $SO_2$  emission into the environment.

In the recent past, the researchers for reducing SO<sub>2</sub> content of stack gases have proposed numerous methods. A critical review on commercial and proposed technologies may be found in a recent pollution technology review [Slack and Hollinden, 1975; England and Beery, 1971]. The techniques actually available for SO<sub>2</sub> removal from flue gases may be broadly classified into wet and dry systems. A number of wet and dry flue gas desulphurization (FGD) processes have been proposed and tested on laboratory and pilot plant scale. Most of the FGD processes tested on commercial scale in developed countries are in the wet category such as (i) limelimestone scrubbing, (ii) double alkali scrubbing, and others, all of which involve a waste disposal problem. So far only three dry recovery FGD processes have reached the commercial stage in the developed countries like USA and Japan, and these include (i) Monsanto's Catalytic-Oxidation process, (ii) Shell's copper oxide regenerative adsorption of SO<sub>2</sub> which produces elemental sulfur and (iii) Mitsubishi's regenerative oxidative adsorption process.

The first commercial use of FGD to control sulfur dioxide pollution from a power plant was in the United Kingdom in the early 1930s. Flue gases were washed with alkaline water from the Thames River with chalk added into it at the Battersea and Bank side Power Stations in London, England. The spent water was discharged back into the Thames after settling and oxidation. This FGD system operated successfully for over 40 years, with 95 percent SO<sub>2</sub> removal efficiency, until power plant closed in 1975. An explosive growth and installation of FGD units however occurred during 1970s in the USA and Japan mainly due to regulatory pressures.

In the early 1960s, major emphasis was laid upon processes for recovery of sulfur in usable form. By 1967, then such processes were being tested or planned in large power plants or other plants that emit sulfur oxides in Germany, England, Japan, and USA. Concurrently, work on dry limestone injection processes was found in the Federal Republic of Germany, Japan, Polland, and USA. In Japan, a number of processes have been tested commercially for simultaneous removal of sulfur and nitrogen oxides, which include activated carbon process, electron beam irradiation,

catalytic reduction with ammonia and wet oxidation-adsorption. High cost for activated carbon and electron beam irradiation or ammonium sulfate formation and equipment plugging are some of the problems which necessitate the desirability of an alternative process for flue gas treatment (FGT). Though the interests in recovery processes continued to avoid waste disposal problem, the throwaway processes were generally considered to be less complex and thus more amenable to short-term development and application.

However, dry FGD processes have received considerable attention in recent years for some specific advantages over other techniques. The process involves contracting flue gas with suitable sorbent which results in dry products for disposal or reuse. The process permits the flue gas treatment at temperatures, which are sufficiently high so that buoyancy of the flue gas is not reduced significantly and reheating can be avoided before the clean flue gas is vented to the atmosphere through stacks. Further, it has been found advantageous to use such dry method for removing simultaneously sulfur dioxide, oxide of nitrogen and particulate matter. Recently dry scrubbing with nahcolite (a naturally occurring sodium bicarbonate mineral) has been claimed to remove approximately 10 to 40 percent of  $NO_x$  and 80 per cent of  $SO_2$  contained in the flue gas.

### 1.2.1 Major wet processes for SO<sub>2</sub> removal

In the wet scrubbing methods, gas mixtures are selectively removed by absorption process with the help of a chemically reactive liquid. Physical absorption involves the dissolution of the gaseous components in the absorbing liquor. If the liquor contains a reactant with which the gaseous component reacts, then the physical absorption is followed by a chemical reaction. Major absorption studies on the selection of absorbents in flue gas desulphurization have been reported by many researchers [Slack and Hollinden, 1975; Gogineni, 1975; Tufte, 1975; Kohl and Reisenfeld, 1985; Meikap and Biswas, 1999]. Some of the major wet processes for SO<sub>2</sub> removal have been outlined below:

(a) **Direct lime/limestone scrubbing**: The lime and limestone scrubbing system uses slurry of lime or limestone to absorb  $SO_2$  from waste gases, thereby reproducing a waste product in the form of a slurry or sludge containing calcium sulfate and calcium sulfite. This method creates a problem for disposal of waste. Further, such systems generally employ some type of scrubbed gas reheat system to avoid condensation and

consequent corrosion of downstream equipment, to suppress the formation of visible plume, and to improve rise and dispersion of stack gas.

(b) **Double alkali scrubbing**: Double alkali systems use scrubbing solutions of soluble alkali salts. The spent scrubbing liquor is then reacted with lime outside the scrubber system, thereby collected  $SO_2$  is precipitated as calcium salts which is purged from the system. The use of clear reactive solution offers the potential for a higher  $SO_2$  absorption rate.

(c) **Magnesium oxide scrubbing**: Magnesium oxide (magnesia) is used to remove  $SO_2$  and magnesium sulfate formed thereby is separated by centrifugation. The magnesium sulfate is then dried and calcined (oxidized by heating) to regenerate magnesium oxide which is reused and the concentrated stream of  $SO_2$  is converted into sulfuric acid.

(d) Zinc oxide process: In this process, the flue gases are contacted with a solution of sodium sulfate and bisulphate whereby  $SO_2$  is absorbed. The solution is next passed into a clarifier, in which particulate matter removed from the gas stream is separated, and finally into a mixer in which it is treated with zinc oxide. After agitation, zinc sulfate precipitate is removed by settling and filtration. Calcining the zinc sulfate results zinc oxide and gas with 70% water and 30%  $SO_2$ .

(e) Wellman-Lord Process: Sodium sulfate is used in this process as the absorbent to obtain sodium bisulphate which is heated to regenerate sodium sulfate and  $SO_2$  in a concentrated form. The recovered  $SO_2$  is further processed for liquid  $SO_2$ , sulfur, or sulfuric acid.

The wet FGD systems, which are considered commercially viable, include direct lime/limestone scrubbing and double alkali scrubbing, while the important recovery FGD systems are magnesium oxide scrubbing and Wellman-Lord process. Many engineering and process problems however are yet to be solved for successful operation of FGD system. Closed loop (no liquid discharge) operation is required if gas cleaning systems are to avoid contributing to water quality problems. The capability of scrubbing system to operate for long periods under closed-loop conditions is still to be demonstrated. Further, flue gas desulphurization by wet system use primarily two types of liquid absorbents viz. slurry or clear liquor, which causes scaling, plugging and corrosion. In addition to these problems, retrofitting of this method requires a large amount of space and high capital cost, especially for wastewater purification.

### 1.2.2 Major dry processes for SO<sub>2</sub> removal

Among the various methods for the removal of  $SO_2$  arising from industrial processes, dry control processes have received considerable attention in recent years due to certain specific advantages over the other conventional techniques or methods. Dry processes would be advantageous since they would avoid problems like flue gas reheating, contacting the gas with a liquid etc., which often pose problems in flue gas wet-scrubbing. In fact, dry  $SO_2$  control system has been considered as one of the most preferred techniques for flue gas cleaning on the grounds of techno-economic aspects besides other advantages. Dry control system typically removes 70 percent of  $SO_2$  in a waste gas stream. It is 15 to 30 percent cheaper to install and operate than a conventional wet scrubbing system. Dry systems usually have savings in reheating and pumping requirements, resulting in 3 to 5 percent savings based on plant operation. Dry  $SO_2$  control system can be advantageously annexed to an existing plant equipped with dry dust collection equipment.

Some of the dry SO<sub>2</sub> control systems are; (i) spray drying, (ii) dry injection, (iii) addition of alkaline reagent to a fuel prior to combustion, and (iv) electron beam irradiation etc. developed particularly in USA and Japan. Out of these systems spray drying has been claimed to be popular and commercially successful. The first commercial sprays dryer-based FGD system was installed at the Strathmore Paper Company in USA for an industrial pulverized coal-fired boiler.

(a) **Spray dryer-based systems**: These systems mainly include reagent feeding and handling equipment; the spray dryer type reactor, and the particulate collection device. The spray dryer can use rotary, two-fluid or nozzle atomization, and the vessel can be anything from the back-mix reactor to a large horizontal duct. In the spray dryer, the atomizers create a spray of fine droplets of alkaline material solution or slurry. When the hot, incoming flue gas at the air pre-heater outlet temperature, (generally 120 to 205 <sup>O</sup>C) contacts the cloud of reagent spray, it rapidly heats the reagent and evaporates the water present. As this occurs, the pollutants in the flue gas react with the solid reagent and available oxygen to form compounds which become dry powder before they impinge upon the reactor walls. Thus no scaling or corrosion can usually occur as the result of wet/dry interfaces. The scrubbed flue gas and particulate matters leaving the spray dryer are separated by the downstream fabric filter or electrostatic precipitator. The choice of fabric filter as the particulate

collection device has an inherent advantage that unreacted sorbent in the collected solids on the filter surface can still react with remaining pollutant in the flue gas.

(b) **Dry injection process**: Dry injection schemes generally involve pneumatically introducing a dry, powdery alkaline material into a flue gas stream with subsequent particulate collection. The injection point has been varied from the boiler furnace area all the way to the flue gas entrance to an electrostatic precipitator or fabric filter (bag collector). Most dry injection schemes use a sodium-based sorbent. Many dry injection programmes in USA have used nahcolite as a sorbent. Nahcolite is a naturally occurring mineral which is about 80 percent sodium bicarbonate. Sodium bicarbonate appears to be more reactive than sodium carbonate. The following overall reaction illustrates this point.

$$2NaHCO_3 + SO_2 = Na_2SO_3 + H_2O + 2CO_2$$
(1)

$$Na_2CO_3 + SO_2 = Na_2SO_3 + CO_2$$
<sup>(2)</sup>

Since bicarbonate loses three moles for every more of  $SO_2$  removal, bicarbonate particles tend to have larger pore volume and are apparently less susceptible to blinding on reaction than sodium carbonate particles.

(c) **Combustion of fuel/sorbent mixture**: The technique involves combustion of coal/alkali mixture and other various dry systems which primarily involve addition of alkaline reagents to a fuel prior to combustion. The SO<sub>2</sub> released from the fuel reacts with the sorbent to form compound which is subsequently collected with the ash. The current research on combustion of fuel/sorbent mixture has taken two forms: (i) combustion of coal/limestone pellets in an industrial stoker-fired boiler, and (ii) combustion of a pulverized coal/alkali fuel mixture in a low-NO<sub>x</sub> burner. The second process is commonly referred to as limestone injection in a multistage burner (LIMB), and efforts to optimize SO<sub>2</sub> and NO<sub>2</sub> removal using LIMB technology are underway.

Preliminary results of bench-scale test work on both processes have indicated that up to 80 percent of the available sulfur in the fuel can be retained by the limestone. The ratio of the calcium to sulfur in the coal/limestone fuel mixture is important in determining how much sulfur is retained. A Ca : S mole ratio 7 : 1 has been used so far, but further work indicted good results with pellets of Ca: S ratio of 3.5 : 1, having mechanical strength, durability and weatherability characteristics comparable to that of raw coal [Kelly and Palazzolo, 1983].

(d) **Electron-beam irradiation**: This technique involves reagent injection into the flue gas, followed by electron-beam (E-beam) irradiation. The E-beam process is in an early developmental stage and has not been applied to a real coal-fired flue gas [Kelly and Palazzolo, 1983].

Of the four dry  $SO_2$  control systems, only spray drying has been commercialized. Dry injection of nahcolite into flue gas has been successfully demonstrated in USA, but unavailability of nahcolite hinders commercialization of this technology. The aforesaid first two systems provide both  $SO_2$  and particulate matter removal, and the last two provide simultaneous  $SO_2$  and  $NO_X$  control. Spray drying demonstration and pilot scale testing and research at present is focusing on refinement of spray dryer design and operation, comparison of rotary and nozzle atomizers, and investigation of the mechanisms and benefits of solid recycle for lime reagent. Other areas of spray drying research and development include optimization of lime slaking and investigation of alternate reagents such as limestone, dolomitic lime, adipic-acid enhanced lime and limestone, and magnesium oxide. Besides the above systems, other systems of sulfur dioxide removal include condensation, oxidation etc.

### 1.2.3 Comparison between dry and wet SO<sub>2</sub> removal processes

The comparison may be made on the basis of the factors such as waste disposal, reagent requirement, operation and maintenance, energy requirements, and economics. Focus has been made on general aspects of dry FGD systems as compared to conventional wet scrubbing systems.

With regard to waste disposal, dry FGD systems have an inherent advantage over conventional wet scrubbing system in that they produce a dry, solid waste product that can be handled by conventional ash handling technique eliminating the requirement for a sludge handling system.

In general, dry FGD systems require a higher stoichiometric ratio of sorbent to inlet  $SO_2$  concentration to achieve the desired removal efficiency than that by conventional wet scrubbing systems. The scaling potential in wet limestone systems however requires extra efforts to maintain proper scrubbing operation.

Dry control systems are less complex than wet systems and offer the prospect of greater reliability and are possibly more flexible with respect to process operations, like flue gas volumetric flow rates and variations in inlet SO<sub>2</sub> concentrations. In general dry systems require less investment cost compared to that in wet systems, which require the thickeners, centrifuges, vacuum filters, and mixers for handling wet sludge waste product.

Further, so far energy requirement for dry FGD systems is concerned, this process have to have economic advantage over wet systems due to savings in reheat and pumping requirements. Many wet FGD systems reheat the flue gas before it enters any downstream equipment to prevent corrosion.

Although costs are quite site specific, one of the major driving forces for development of dry  $SO_2$  control systems is the opportunity for reduction in both capital and operating costs. The minimal equipment and operating requirements for dry systems make the process economically attractive so far as capital costs are concerned.

In summary, dry systems do offer potential advantages over wet systems particularly in the areas of energy savings and costs. In spite of crucial issues such as waste disposal problems such systems usually have less problems with respect to corrosion, erosion, plugging and scaling.

### 1.3 Sorbents for dry SO<sub>2</sub> removal processes

Adsorption of SO<sub>2</sub> greatly depends on the nature of sorbent and its reactivity with pollutant gas. Usually sodium based or calcium based sorbents are used. In spray dryer based systems, sodium carbonate solutions and lime slurries are common sorbents. Lime, however, has become the sorbent of choice in many circumstances, particularly in throwaway processes, because of cost advantage over sodium carbonate. On the other hand, most dry injection systems use sodium based sorbent. Nahcolite (a naturally occurring sodium bicarbonate mineral) has been used as a sorbent for many dry injection programmes in USA. In general, it is desired that the sorbent should have the potential for providing a good compromise between reactivity, cost and availability. Major restraints to the development of commercial dry injection/particulate collection systems have been uncertainty regarding acceptable disposal practices for sodium-based waste products. Considering all the aspect, lime is considered to be cost-effective sorbent for SO<sub>2</sub> removal.

It appears that the reactions of  $SO_2$  with lime, magnesia, alkaline alumina, and other reagents are likely to generate a great deal of interest in the forthcoming years, because of their potential role in abating  $SO_2$  pollutions from gaseous effluents. At present the interest in dry processes lies with respect to sorption of SO<sub>2</sub> by lime, using fluidized or injection method

### 1.4 Problems in dry SO<sub>2</sub> removal processes

The limitations of dry sulfur dioxide removal processes are discussed below;

The lime or limestone injection Process [Slack and Hollinden, 1975; Kohl and Riesnfeld, 1985] is one of the simplest systems devised for removal of SO<sub>2</sub>. In this process finely ground limestone or lime is added to the coal or injected into the furnace. The experiments have shown that  $Ca(OH)_2$  shows better utilization. The SO<sub>2</sub> removal efficiency is 30-75% only. Sodium additives enhance the performance of calcium sorbent by modifying the diffusivity of the CaSO<sub>4</sub> product layer.

Lygfelt and Leckner (1989) investigating the performance of a 16 MW circulating fluidized bed boiler and reported that re-emission of SO<sub>2</sub> occurred from SO<sub>2</sub> capture lime due to reduction of CaSO<sub>4</sub> with combustion intermediates, such as CO, H<sub>2</sub> in the dense phase of the bed, at a temperature of 880-890  $^{\circ}$ C and at an excess air ratio of 1.4 at 930  $^{\circ}$ C. The amount of sulfur leaving the boiler as SO<sub>2</sub> was found to be more than double the amount of sulfur added to the boiler in the form of sulfur.

Skrifvars (1991) reported that the limestone injected into the furnace of a coal fired boiler, for the purpose of removing sulfur from flue gas, could cause fouling of the heat exchanger surface as it passes trough the super heater area in the flue gas channel. However, the furnace injection processes are not found to be commercially successful. The process is also not fully characterized for potential negative boiler impacts. Another Japanese process employs manganese oxide and is used by Mitsubishi Heavy Industries. This is a dry sorption process utilizing an entrainment reactor at about 150 °C. The solids are separated from the gas stream by cyclone and an electrostatic precipitator and then subjected to a wet regeneration involving solution of manganese sulfate in water and precipitation of hydrated manganese oxide by alumina and air.

The method of condensation for the removal of sulfur dioxide [Liptak, 1974] requires temperature lower than the saturation temperature of sulfur dioxide (-123  $^{\circ}$ C).Thus the technique of condensation involves a prohibitive cost of refrigeration. Furthermore, the necessity of removing of CO<sub>2</sub> which solidifies at a temperature of - 99  $^{\circ}$ C in the condenser makes the method economically non-viable.

The method of oxidation for removal of sulfur dioxide requires higher concentration of sulfur dioxide gas. But the typical sulfur dioxide gas content in the flue gas in most of the coal thermal power plants, sulfuric acid plants and oil refineries is less than 1000 ppm, which is too low for profitable recovery of sulfuric acid. Innovative dry adsorption technologies such as furnace and post furnace injection techniques have been developed to facilitate burning of high and low sulfur coals in the coal fired thermal power plants, which are available widely on a commercial basis. However the drawbacks of this injection technique are low efficiencies.

But limestone becomes less attractive, because of low level of limestone utilization. Moreover, the principal impurity is magnesium carbonate which results in water soluble product (magnesium sulfite) in the process of sulfur dioxide removal and thereby poses water pollution problem. Another difficulty with limestone is the short residence time (1 or 2 seconds) of the solid at the temperatures at which important reactions like calcium carbonate calcinations and calcium sulfate formation take place. Since gas temperatures in the furnace of a coal-fired power plant boiler are typically above 1040  $^{0}$ C, sorption of SO<sub>2</sub> takes place only after the sorbent has left the furnace and entered the cooler sections of the boiler. Therefore, sorption of SO<sub>2</sub> by lime must take place in the short span of time the sorbent spends between 1000  $^{\circ}$ C and 750  $^{\circ}$ C (where CO<sub>2</sub> sorption presumably provides a competing reaction).

It appears that the furnace injection, post furnace injection, spray drier absorption have low sulfur dioxide removal efficiencies. Therefore, the interest lies on the development of an appropriate reactor which can enhance the removal efficiency and additive utilization.

#### 1.5 Equipment type for sulfur dioxide removal

Equipment for dry systems includes conventional reactor types such as (i) fixed bed, (ii) moving bed, (iii) fluidized bed, and (iv) transport line reactor etc.. Fig. 1.1 presents some of the selected types of contacting for gas-solid reaction. The fixed bed is unsuited for continuous operation and batch operations yield non-uniform product. The moving bed requires fairly uniform sized feed with little or no fines. Adsorption of sulfur dioxide in packed bed has disadvantages of low mass-transfer



Fig. 1. 1. Selected type of contacting for reacting gas-solid systems

coefficient and periodic operation with low treatment capacity. Fluidized bed partially overcomes these problems [Gidaspow, 1994]. Fluidization of bed increases the mass transfer coefficient and therefore, treatment capacity [Kishore and Varma, 2005]. Fluidized beds operating in bubbling and turbulent fluidization regime, mixing of solids and gas by-passing result in poor performance than other type reactors. But the high conversion is possible when the fluidized bed operates in fast fluidization and co-current pneumatic transport regime at high temperature [Kuni and levenspiel, 1991]. Table 1.3 presents the flue gas temperature of targeted industries. It may be concluded that the removal of sulfur dioxide gas in a fluidized bed at low temperature may be a possible solution.

Table 1. 3. Flue gas temperatures in stack

Industry	Temperature of stack gas	
Copper, lead and zinc Smelting (Off gases must be utilized for acid manufacture)	75-85 <sup>o</sup> C	
Sulfuric acid plant	75-85 <sup>o</sup> C	
Thermal power plant	120-140 <sup>o</sup> C	

### 1.6 Critical appraisal of sulfur dioxide removal technologies

Literature review indicates that most of the present day sulfur dioxide emission control systems have been evolved in response to a particular set of environmental regulations and these systems are inflexible and have almost become less economical to operate with the increasing stringency of old regulations or promulgation of new regulations. Options are also limited in using process modifications and raw material substitutions as the methods of pollution control abatement. For example, regulation mandating additional decrease in SO<sub>2</sub> content from flue gases from existing industries would be satisfied by either with additional control equipment at the expense of increased capital cost or reduced capacity. Furnace injection in case of coal-fired thermal power plants leads to corrosion and abrasion of the furnace internals, which require exotic materials of construction, resulting in a costly system. The wet method can cause scaling, plugging and erosion of the scrubbing system. In addition to these problems, the wet sludge/waste water generation creates more problems to handle and dispose of. Therefore, the emission of
sulfur dioxide may be controlled by dry processes in a fluidized bed reactor which create fewer corrosion and scaling difficulties and produce waste product much easier to handle and dispose of. Thus with increased stringent regulations, a cost-effective with high efficiency control technique seems to be only viable alternative to achieve techno-enviro-economic feasibility.

# 1.7 Status of research on removal of sulfur dioxide in fluidized bed reactor

The high mass transfer, good mixing characteristics along with other inherent advantages of fluidized bed reactor have motivated few researchers to investigate on the reactor for removal of sulfur dioxide, which are discussed below;

Kato et al. (1994) reported the use of SO<sub>2</sub> treatment on a lime powder-particle single stage fluidized bed. These researchers treated SO<sub>2</sub> on a bench scale with the addition of limestone, using thick sand particles with diameters of 495–991  $\mu$ m, and fine limestone particles with 2, 5 and 9.9  $\mu$ m diameters. They investigated the influence of temperature (650–950 <sup>O</sup>C), molar ratio of Ca/S (1, 2 and 3), the superficial velocity of the fluidization gas (1.0, 1.5, 2.0 and 2.5 m/s), the height of the initially static bed (10.0, 20.0 and 30.0 cm), and the inflow concentration of SO<sub>2</sub> (500, 700 and 1000 ppm) on removal efficiency. These researchers found that the SO<sub>2</sub> removal efficiency was considerably dependent on the variables investigated , achieving 100% removal at a temperature of 800 <sup>O</sup>C, with a limestone particle diameter of 5  $\mu$ m, Ca/S molar of 2.5, static bed height of 10.0 cm, superficial velocity of 1.0 m/s, and inflow gas concentration of 1000 ppm. Further it was reported that slightly change in operating condition reduced the efficiency appreciably.

Operating the similar system as reported by Kato et al. (1994) but using 29  $\mu$ m dolomitic limestone particles and a (Ca + Mg)/S molar ratio of 2, Tashimo et al. (1998) achieved a removal efficiency of approximately 50% at a temperature of 800  $^{\circ}$ C, a velocity of 1.0 m/s, static bed height of 10.0 cm and in-flow gas concentration of 1000 ppm, as opposed to the 65% achieved by Kato et al. (1994). Jiang et al. (1995) evaluated sulfur dioxide removal from flue gas in a circulating fluidized bed absorber by use of CaO sorbent at humid condition. They reported that SO<sub>2</sub> removal efficiency is a strong function of reactor solids loading and the degree of saturation. Chiang et al. (2003) reported on the application of single stage fluidized bed adsorber integrated with fabric filter for removal of acidic gases from flue gas in an incinerator. The removal efficiency of the adsorber for SO<sub>2</sub> was 48.8-73.5% and it reached over

94% after passing through the filter. Pisani et al. (2003) reported on the application of a continuously operated binary fluidized bed single stage reactor using dolomitic lime stone (24  $\mu$ m) and inert solids of 500-590  $\mu$ m diameters for removal of SO<sub>2</sub>. The maximum gas removal fraction of 76% was achieved at a temperature of 800 <sup>o</sup>C, Ca/S ratio of 3 and velocity of 0.8 m/s. Pisani et al. (2004) reported on the application of a continuously operated binary fluidized bed single stage reactor using dolomitic lime stone (9.1  $\mu$ m) and inert solids of 500-590  $\mu$ m diameters for removal of SO<sub>2</sub>. The maximum efficiency of 97.7% was achieved at a temperature of 700 <sup>o</sup>C with a Ca/S ratio of 3 and a velocity of 0.8 m/s. However, it was reported that slightly change in operating condition reduced the efficiency substantially.

Chu and Hwang (2005) reported the application of a internal circulating fluidized bed reactor using calcium sorbent (385  $\mu$ m) and inert solids of 438  $\mu$ m diameter for removal of SO<sub>2</sub>. The maximum efficiency of 100% was achieved at a temperature of around 30 <sup>o</sup>C at higher relative humidity and at initial concentration of 500 ppm. Chang-Keun et al. (2007) used two stage fluidized-bed reactors in the continuous solid circulation mode to investigate the feasibility of using potassium carbonate-based solid sorbent to remove air pollutants including carbon dioxide. But it was reported that the removal efficiency was very low.

Literature survey reveals that most of studies have been undertaken for removal of  $SO_2$  in single stage fluidized bed reactor at high temperature (above 500  $^{\circ}$ C) contrary to stack gas temperature after ESP in thermal power plant (120  $^{\circ}$ C) and sulfuric acid plant (75  $^{\circ}$ C), since the removal efficiency of sulfur dioxide at low temperature in a single stage fluidized bed reactor is quite low.

# 1.8 Scope and objective of the present investigation

Critical appraisal of the status of the research on the control of sulfur dioxide shows that all the dry process techniques are mostly used in either fixed bed reactor or a single stage fluidized bed reactor at high temperature, which is not suitable for removal of sulfur dioxide from stack gases in the industries. Further, the quantity of  $SO_2$  generated from coal based thermal power plants in India is being discharged to the atmosphere through high stack considering dilution as control of pollution. Development of  $SO_2$  emission standard is on the process by the government's regulating agency and is expected to have stringent emission standard. The other plants such as sulfuric acid plant, oil refineries have wet scrubbing method resulting in generation of liquid effluent, which create more problems to handle with and dispose of. Thus the dry process to control  $SO_2$  at lower temperature is the need of the day and the equipment to be selected for the control of sulfur dioxide must have a very high efficiency of collection.

Literatures suggest that the fluidized bed reactor operating at various regimes can be used as possible equipment for removal of sulfur dioxide at high temperature. But, at low temperature the efficiency of the reactors is very low besides other limitations. The limitations of a single stage continuous fluidized bed reactor can be avoided by the use of multistage continuous fluidized bed reactor due to its staging effects [Miracca and Capone, 2001]. Due to the counter current operation, the effective concentration gradient is relatively higher which, in turn, results in the enhanced separation efficiency. The gas and solid flow rates may be varied over a wide range in the column as per the requirement. Further, the direct literature is very limited for removal of  $SO_2$  in multistage fluidized bed reactor.

# 1.9 The plan of investigations

In view of reported literature on non-suitability of single stage fluidized bed reactor for removal of sulfur dioxide from flue gas at low temperature, multistage fluidized bed reactor seems to be an alternative option for the purpose. So the major objectives of the present investigations include;

- (1) Design and development of a multi-stage fluidized bed reactor for control of gaseous pollutants.
- (2) Studies on the stable operating range of the multistage fluidized bed reactor and identification of fluidization regimes.
- (3) Hydrodynamics of the multistage fluidized bed reactor for single particle system such as hydrated lime, sand particles for the measurement of pressure drop, emulsion phase hold-up and mean solids holding time.
- (4) Studies on dynamics of downcomer in multistage fluidized bed reactor.
- (5) Hydrodynamic characteristics such as pressure drop of the multistage fluidized bed reactor for mixed particle system (lime and dolomite mixture).
- (6) Performance studies on removal of SO<sub>2</sub> by hydrated lime particles in the multistage fluidized bed reactor.
- (7) Theoretical analysis on multi-stage fluidized bed reactor for prediction of removal efficiency of SO<sub>2</sub>.

# **CHAPTER 2**

# DESIGN, DEVELOPMENT AND CHARACTERIZATION OF A MULTISTAGE FLUIDIZED BED REACTOR

# Design, development and characterization of a multi-stage fluidized bed reactor

### 2.0 Introduction

"The arrival time of a space probe traveling to Saturn can be predicted more accurately than the behavior of a fluidized bed chemical reactor". Even though the above quotation of Geldart (1986) is almost 22 years old it remains true in the new millennium of fluidization engineering. In spite of all complexities and ambiguities, fluidized bed is one of the best-known contacting methods used in the chemical process industries. Among its chief advantages are that the particles are well mixed leading to low temperature gradients, they are suitable for both small and large scale operations and they allow continuous processing. There are many well established operations that utilize this technology, including cracking and reforming of hydrocarbons, coal carbonization and gasification, ore roasting, Fischer Trospch synthesis, coking, aluminum production, melamine production, and coating preparations. The application of fluidization is also well recognized in nuclear engineering as a unit operation for example, in uranium extraction, nuclear fuel fabrication, reprocessing of fuel and waste disposal.

Despite the serious drawbacks, the compelling advantages of overall economy of the fluidized contacting system have been responsible for its successful use in many industrial operations. Understanding the deficiencies of fluidized bed contacting devices and an effort to overcome them can lead to successful operation of difficult systems.

# 2.1 Fluidization regimes

The gas-solid fluidization regime has been extensively studied by many researchers [Yerushalmi and Cankurt, 1979; Simone and Harriott, 1980; Kwauk and Li, 1996; Zijerveld et al., 1998; Smolders and Baeyens, 2001]. When the solid particles are fluidized, the fluidized bed behaves differently as velocity, gas and solid properties are varied. It has become evident that there are number of regimes of fluidization, as shown in Fig. 2.1. When the flow of a gas passed through a bed of

particles is increased continually, a few vibrate, but still within the same height as the bed at rest. This is called a fixed bed. With increasing gas velocity, a point is reached where the drag force imparted by the upward moving gas equals the weight of the particles, and the voidage of the bed increases slightly: this is the onset of fluidization and is called minimum fluidization with a corresponding minimum fluidization velocity, u<sub>mf</sub>. Increasing the gas flow further, the formation of fluidization bubbles sets in. At this point, a bubbling fluidized bed occurs. As the velocity is increased further, the bubbles in a bubbling fluidized bed will coalesce and grow as they rise. If the ratio of the height to the diameter of the bed is high enough, the size of bubbles may become almost the same as diameter of the bed. This is called slugging. If the particles are fluidized at a high enough gas flow rate, the velocity exceeds the terminal velocity of the particles. The upper surface of the bed disappears and, instead of bubbles, one observes a turbulent motion of solid clusters and voids of gas of various sizes and shapes. Beds under these conditions are called turbulent beds. With further increase in gas velocity, eventually the fluidized bed becomes an entrained bed in which we have disperse, dilute or lean phase fluidized bed, which amounts to pneumatic transport of solids.



Changes in Regime with increasing in Gas Velocity.

### Fig. 2. 1. Change in regimes with increase in gas velocity

Monazam et al. (2005) conducted a series of experiments and took two distinct transition velocities; the lowest transition velocity marked the transition between the

dense-phase turbulent and the fast fluidization flow regimes, while a higher or second transition represented the transition between the fast fluidization and the dilute-phase flow regimes, to identify different regimes. However the regimes described by Yerushalmi and Cankurt (1979) are frequently used to describe the regime of fluidization.

### 2.2 Geldart's classic classification of powders

The behavior of solid particles in fluidized beds depends mostly on their size and density. A careful observation by Geldart (1973) is shown in Fig. 2.2 in which the characteristics of the four different powder types were categorized as follows:

• Group A is designated as 'aeratable' particles. These materials have small mean particle size ( $d_p < 30 \mu m$ ) and/or low particle density (<1.4 g/cm<sup>3</sup>). Fluid cracking catalysts typically are in this category. These solids fluidize easily, with smooth fluidization at low gas velocities without the formation of bubbles. At higher gas velocity, a point is eventually reached when bubbles start to form and the minimum bubbling velocity,  $u_{mb}$  is always greater than  $u_{mf}$ 

• Group B is called 'sandlike' particles and some call it bubbly particles. Most particles of this group have size 150  $\mu$ m to 500  $\mu$ m and density from 1.4 to 4 g/cm<sup>3</sup>. For these particles, once the minimum fluidization velocity is exceeded, the excess gas appears in the form of bubbles. Bubbles in a bed of group B particles can grow to a large size. Typically used group B materials are glass beads (ballotini) and coarse sand.

• Group C materials are 'cohesive', or very fine powders. Their sizes are usually less than 30  $\mu$ m, and they are extremely difficult to fluidize because interparticle forces are relatively large, compared to those resulting from the action of gas. In small diameter beds, group C particles easily give rise to channeling. Examples of group C materials are talc, flour and starch.

• Group D is called 'spoutable' and the materials are either very large or very dense. They are difficult to fluidize in deep beds. Unlike group B particles, as velocity increases, a jet can be formed in the bed and material may then be blown out with the jet in a spouting motion. If the gas distribution is uneven, spouting behavior and severe channeling can be expected. Roasting coffee beans, lead shot and some roasting metal ores are examples of group D materials. Though Geldart's classification of solids is used most widely, different approaches have also been presented to define



A: Aeratable (u<sub>mb</sub> > u<sub>mf</sub>) B: Bubbles above u<sub>mf</sub> (u<sub>mb</sub>= u<sub>mf</sub>) C: Cohesive D: Spoutable

Fig. 2. 2. Geldart classifications of particles for air at ambient conditions (Geldart, 1973)

the boundaries between groups of particles such as A/B boundary, A/C boundary, B/D boundary. Molerus (1982) has attempted to distinguish between group A and C by considering the balance between hydrodynamic and cohesive forces. It has been suggested that the criterion for powder eligible to group C if

$$\frac{10(\rho_s - \rho_g)d_p^3 g}{F_H} < 10^{-2}$$
(2.1)

where,  $F_{\rm H}$  is the adhesion force transmitted on a single contact between two adjacent particles.

Geldart's classification is clear and easy to use as displayed in Fig. 2.2 for fluidization at ambient conditions and for u less than about  $10 \cdot u_{mf.}$  For any solid of a known density  $\rho_s$  and mean particle size  $d_p$  this graph shows the type of fluidization to be expected. It also helps predicting other properties such as bubble size, bubble velocity, the existence of slugs etc.

# 2.3 Bubbling fluidized beds

Fluidization of particles in bubbling regime is possible with gas velocities ranging from minimum fluidization velocity to terminal settling velocity of particles. When, the fluidized bed operates in bubbling fluidized bed type as shown in Fig. 2.1, gas fluidized beds are characterized by the 'bubbles' which form at superficial gas

velocities only slightly higher than that required to just fluidize the particles [Chi and Wang, 2002]. This type of fluidization has been called 'aggregative fluidization'.

Any continuous fluidized systems operating in bubbling or any regime have different residence time for solid particles yielding non-uniform product. The limitations of a single stage continuous bubbling fluidized bed reactor can be avoided by the use of multistage continuous fluidized bed reactor due to its staging effects. Due to the counter current operation, the effective concentration gradient is relatively higher which, in turn, results in the enhanced separation efficiency. However minimum fluidization velocity and terminal velocity differ in a multi-particle system in a multistage fluidized bed from that of a conventional fluidized bed, as multistage fluidized bed is susceptible to flooding. Before a detail on multistage fluidized bed is investigated, various types and operation of bubbling fluidized bed reactors need to be discussed. Bubbling fluidized beds can be operated as batch or continuous mode.

#### 2.3.1 Batch fluidized bed reactor

In batch fluidized beds, the particles position is defined as a function of time and the properties at any point of the bed are same. The only major drawback is that the batch systems are not suitable for large-scale operation. Further, batch fluidized beds can be operated in single stage as well as multistage.

## Single stage batch fluidized bed reactor

In single stage batch fluidized systems, solids are handled as batch and gas is continuously passed through the bed. The time of operation is usually governed by the system requirements. The contact of the gas with solids is once through then the quality of solid product is uniform. The efficiency of operation especially with gas phase will be low. Depending on the gas velocity, the system will be either in fixed bed state or at incipient fluidized state or fluidized state or elutriation.

#### Multistage batch fluidized bed reactor

In multistage batch systems, the gas moves from stage to stage continuously coming in contact with solids in each stage, which are handled as batches. Two alternate methods of contacting solids and gas in multi-stage batch systems are shown in Fig. 2.3 and 2.4 [Rao and Gopalkrishna, 1975]. A single column is divided into a number of individual stages as shown in Fig. 2.3 with each stage fitted over the other.

The solids are fed as batches to each stage at point A and can be discharged after the operation either from the same point or at point B. The gas enters at the bottom of the column at C and leaves at point D after contacting on the solids in each stage. The unit is compact and requires less floor space. Fig. 2.4 shows a multi-stage batch unit where each stage is a separate unit and the gas entering at first stage passes in series through all stages before its exit. The solids in each stage are handled as batches.

The major draw back, in batch process is the considerable time loss due to charging and discharging of solids as batches in each stage. This is a limitation on its adaptability in large scale operation.



Fig. 2. 3. Multistage batch fluidization units (One unit)



Fig. 2. 4. Multistage batch fluidization units (Multiple units)

In multistage batch units for gas solid contacting, the solids pass through first in fixed bed stage at lower gas velocities and then experience the onset of fluidization with increase in gas flow rate and enter the fluidized state as in a single stage unit. Thus it is expected that all the parameters which govern the behaviour of the single stage batch fluidizing systems are also applicable to multistage batch units. The major difference between the two is the lower bed height of solids in each stage of a multistage batch unit and series contact of gas with solids in multistage unit.

The pressure drop in multistage units is much higher than in identical single stage batch unit due to the presence of larger number of grids. The single stage batch units have more chances of slugging due to the use of deeper beds. This is generally avoided in multi-stage batch unit by the use of shallow beds. This is generally avoided in multi-stage batch unit by the use of shallow beds. Use of shallow beds normally leads to better quality of fluidization. For shallow beds it is recommended [Vanecek et al., 1966] that the grid resistance should be of the same or larger than the resistance of the bed. The grid resistance should be such that even in case where a section of the grid is free, the velocity of the gas in the layer of material in the rest of the grid should not fall below the fluidizing point. Such a resistance will ensure that fluidized bed will be renewed over the whole area and chances of channeling are avoided.

Bed expansion characteristics are of importance in multistage batch units, as they dictate the order of tray spacing. At higher gas velocity, the bed expansions are large and the grid spacing should be large to ensure that solids of the stage below do not touch the grid; otherwise the performance of the column is adversely affected. Large tray spacing will lead to higher vertical space requirement. This can be limited either by the use of lower fluid velocity or use of shallow beds. The former is governed by the system requirements and usually shallow beds are made use of to limit the overall height of the column.

Batch fluidized beds, both single and multistage, are not much favored due to their limitations for use in large scale operation which requires very large unit. This is over come by the use of continuous fluidizing systems.

# 2.3.2 Continuous fluidized bed reactor

In continuous fluidization, the solids enter continuously from one end and get discharged at the other end while they come in contact with continuously flowing fluid medium during the transit. The time of stay of the solid in the bed or the bed hold up can be controlled. The continuous fluidizing systems can be single stage or multistage units.

The behavior of single stage continuous fluidizer is identical to that of batch fluidized bed with regard to minimum fluidizing velocity, bed pressure drop, bubbling and slugging phenomena and bed expansion. Sun and Grace (1992), Grace and Sun (1991) have studied particle size distribution in bubbling fluidization regime for continuous systems. In continuous systems, the bed performance is related to the time of contact between solids and gas. Thus, the residence time distribution (RTD) of solids and gases and their hold-up is of significance in designing continuous contacting devices.

Danckweerts at al. (1954), May (1959), Wen and Yu (1966a), Pell (1990), Sundarajan and Krishnaiah (1998) and many other workers have studied the flow pattern of gases in continuous single and multistage fluidization. Namkoong et al. (1966) and Gauthier and Flamant (1991) conducted dynamic response studies to determine gas RTD. Based on the study the nature of flow of gas as plug, back mix or arbitrary flow is determined. Experimental results indicate that the gas flow in fluidized beds lies squarely between the two extremes of plug flow and back-mix flow. But, when the number of mixing stages is larger, plug flow is approached. For practical applications it is not enough to know how long the fluid element stays in the bed, but its history must be known whether it slipped through the bed as a part of a bubble or whether it spent most of its time percolating through the emulsion phase. For determining this, stimulus response studies have been conducted using different models [Levenspiel, 1962].

## 2.4 Continuous multi stage fluidization

The limitations of a single stage continuous fluidizer can be summed up in terms of wide distribution of residence time of solids and low efficiency of operation both with respect to gas and solid phase besides the chances of slugging in deep beds. The baffles are used to partition the reactor to overcome certain disadvantages associated with single-stage fluidization (Verma, 1975; Pillay and Varma, 1983) .The provision of internal baffles not only facilitates the continuous co-current or counter-current flow of the phases at near plug flow condition, but also establishes the

temperature and concentration gradients along the length of the bed, in addition to limiting the formation and growth of bubbles. Besides these, this also offers other advantages such as reduction in axial mixing of phases, higher transfer rates, reduction in size of equipment.

Continuous multistage contacting of gas and solids can be obtained by the following methods :(i) Simple multiple contacting system. (Fig. 2.5) (ii) Cross flow contacting system (Fig. 2.6) (iii) counter current contacting system (Fig. 2.7).



Fig. 2. 6. Multistage continuous cross-flow fluidizer

In multistage continuous systems with simple multiple contacting as in Fig. 2.5, solids are fed continuously at point A located at different heights and product from each stage is collected at point B. The gas moves from stage to stage from bottom and its condition differs substantially as it moves up. There is no inter-stage transfer of solids and solid product quality varies from stage to stage and it varies even in one stage due to wide variation in the residence time distribution of solids.

Better product quality can be achieved by increasing the solid residence time and narrowing its distribution as in cross flow or counter current multi-stage units. In cross flow units as shown in Fig. 2.6, solids are fed at point A and move from chamber to chamber and finally get discharged from the unit at point B. Gas is fed separately to each chamber at point C and has only once through contact with the solids. Thus the requirements of the fluidizing medium are large and efficiency of operation with respect to gas phase will be low. As far as the solids are concerned due to their multiple contacting with the gas, the residence time distribution is more uniform giving better quality product. However, slugging may occur if deeper beds are used.

The counter current multistage system is an improvement over the cross flow system. Here the solids enter at the top and move down from section to section through the down comers and finally leave from the bottom. The gas enters at the bottom and moves upward contacting the solids in each stage and finally leaves at the top. These units resemble a plate type distillation column. Due to the multiple contacting the solid product is more uniform. The residence time distribution has a definite improvement over the single stage unit. The gas requirements are less than in a cross flow unit. Thus the unit gives better thermal and conversion efficiency for both gas and solid phase.

The behavior of the multistage counter current fluidizer will be similar to single stage continuous fluidizer with respect to minimum fluidizing velocity, bed pressure drop in each stage. The difference will be due to increase in number of stages which give rise to increased pressure drop. The bed expansion is of significance in multi-stage units as it governs the grid spacing. Too low a spacing will lead to contacting of the solids from stage below with the solids on any grid thus completely jeopardizing the counter current behavior. The beds are usually small in counter current units and hence chances of slugging are less. But for proper solid distribution on the tray, it is essential to design the grids properly. The flow pattern of solids and gases tend to be plug slow with counter current behavior. The residence time distribution of solids evens out with increase in number of stages. The efficiency of operation of multistage processes is higher than those in single stage process [Vanecek et al., 1966] The efficiency approaches unity as the number of stages are increased.

Because of the advantages mentioned, multistage units have been accepted in industries. Multistages are being units used for various applications involving heat transfer [Pevel at al., 1966; Toei and Akao, 1968], Mass Transfer [Etherington et al., 1956; Ermenc, 1961; Rowson, 1963], reactions like Ore reduction and beneficiation [Beeken, 1960; Zenz and Othmer, 1960; Boucrant and Imretoth, 1966; Ketteridge, 1965; Legler, 1970; Kilian and Dickey, 1970; Zhenfu et al., 2007]. Almost every industrial application is preceded by an extensive and laborious pilot plant study. This has been due to lack of understanding of the basic mechanisms of transfer of solids from stage to stage and the process of fixing number of stages and prediction of bed behavior. In case of multistage continuous fluidizations, practice seems to be far ahead of theory.

The efficiency of operation of a multistage process is always higher than a single stage process. Number of theoretical stages can be calculated based on efficiency [Kuni and Levenspiel, 1991; Vanecek et al., 1966] and process driving force and RTD. Kuni and Levenspiel (1991), Streltsov and Streltsov (1972a and 1972b) have calculated analytically the number of stages required for counter current operation for single size and poly disperse solids phase based on kinetic data assuming well mixed back mix flow for solids and plug flow for gas. Akopyan et al. (1967) have derived empirical equation based on process driving force for mass transfer operations for calculating what they called "Pseudo-sections" using the concept of degree of utilization of the driving force. Moldov and Ishkin (1967) conducted adsorption experiments in a five stage column and compared the performance with the theoretical analysis based on the concept of NTU. The theoretical concept agreed quite well with experimental results. Todes and Lezin (1967) developed algorithms for computation of number of stages for adsorption and de-adsorption. Prediction of number of stages based on heat transfer has been attempted by Pevel et al., 1966. In spite of the above reported literature, there is no standardized method for estimating the number of stages of a multistage counter current unit. In practice the actual number of stages is seldom more than five or six. These may be due to difficult solids down flow prevalent in a multistage column.

These multistage fluidized beds can be categorized basing on the transfer of the solids from one stage to next stage below; (a) solids passing through perforated plates, (b) solids passing through downcomer which are simply empty tubes allowing the transfer of solids from upper fluidized bed to lower one (Martin-Gullon et al., 1995 and 1996). In the former the diameter of perforated plate holes are large enough to allow simultaneous flow of solids and gas through them. Enough literature is available with studies carried out on the multi-staged fluidization without downcomer. On the other hand, the literature of the stage-wise operation of the gas–solid reactor with downcomer is very insufficient.

Therefore, in the present study, a multistage counter-current gas-solid fluidized bed reactor operating in three stages has been designed and constructed so that the hydrodynamic as well as the mass transfer characteristics could be improved. An attempt has therefore been made to acquire knowledge on the various components and their characteristics and effect in the system.

#### 2.4.1 Components of multistage countercurrent fluidized bed reactor

The major components such as downcomer and distributor are expected to play major role in stable operation of multistage fluidized bed reactor. The investigations made by few researchers on these components are outlined below;

#### Downcomer

The overflow standpipe, or downcomer, is simply an empty tube which permits the transfer of solids from an upper fluidized bed to a lower one, where the standpipe is sunk in the lower bed. These are usually located on the periphery and should be filled constantly with solids to prevent the flow of gas through them in large quantities [Yagi and Muchi, 1955; Rose et al., 1962]. External control of solid levels in downcomer will be difficult. To prevent or to reduce fluid up-flow in down comers, a section of grid below the downcomer may be covered by a cap. Alternately caps or flaps may be fixed to the end of the downcomer. The feeding of solids into a fluidized bed can be either by pneumatic conveyance or by gravity flow. The gravity flow through long downcomer gives better solid mixing characteristics and less of solid by passing as reported by Singer et al. (1957). The use of dip legs (downcomer) poses problem when the unit is started up [Davidson and Harrison, 1963]. Since the inter stage transfer of solids is difficult by the downcomers due to the pressure differential between the two stages, mechanical devices like externally controlled valves [Vanecek et al., 1966] on the downcomers reported to have been used.

Vanecek et al. (1966) suggested use of conical downcomer tubes for maintaining stable operating conditions. They suggested the use of conical down comers with increasing or decreasing cross section from top to bottom with a flap at the bottom. It is recommended that the narrow end diameter of the downcomer through which the solids flow should be at least six times the largest particle size in a poly disperse mixture. The top end of the downcomer is recommended to be three times the bottom end.

Stemerding et al. (1963) discussed a method of introduction secondary air into a cup below the down comers to maintain smooth operating conditions. Levinson et al. (1967) mentioned about four different types of arrangements used below the down comers for liquid fluidized beds to ensure smooth flow. Molodov and Ishkin (1966) studied a five-stage fluidized bed adsorber, with the stages connected by overflow standpipes, for a solid mass flux in the downcomer of around 1.0 kg/m<sup>2</sup> s. These authors pointed out that for a stable operation the solids in the overflow standpipes must be unfluidized, and constrictions were therefore placed at the bottom of the downcomers. They assumed that the gas velocities through the fluidized beds and through the downcomer constriction are equal.

Papadatos et al.(1975) studied the solids holdup in a multistage fluidized bed system operating in continuous regime with activated carbon, varying the solids feed rate and the downcomer diameter. These authors proposed a relationship to obtain the solid holdup in a fluidized bed as a function of the solids feed rate, gas velocity, particle size, weir height, weir diameter and column diameter.

Later, Eleftheriades and Judd (1978) carried out a study of a two-stage fluidized bed system with downcomers (with no constrictions), using sand (mass flux from 10 to 300 kg/m<sup>2</sup>·s). Their objective was to study the gas velocity range in which the solid regime in the standpipe was a moving bed, varying both gas velocity and solid flow rate. When the gas velocity (through the overall system) is close to the minimum fluidization velocity, the solid mass flux necessary for moving bed regime is nearly 30 kg/m<sup>2</sup>· s. If the gas velocity was increased, the solid mass flux also had to be increased to maintain the moving bed regime, until a maximum is reached, and at

this point the solids regime changed to fluidized bed in the downcomer (around 200  $kg/m^2$ ·s). This maximum varied slightly with the dimensions of the downcomer.

They also explained the various regimes of flow in downcomer. These are: (a) In the slip-stick flow regime which is characteristic of completely non-aerated flow of large particles. In this regime the solids move downwards in a jerky fashion. (b) In moving bed flow regime where the solids flow smoothly downwards with a small relative motion between particles and in absence of significant discontinuities such as cracks or bubbles. The voidage is little smaller than the voidage at incipient fluidization.

(c) In the fluidized bed flow regime where the flow pattern resembles a fluidized bed of solids traveling down the downcomer. Bubbles are present, and the overall voidage in the downcomer is greater than the minimum fluidization voidage.

(d) In the dilute phase regime where the particles are streaming down either singly or in small clusters and the overall voidage is almost unity.

According to Eleftheriades and Judd (1978), the Molodov and Ishkin (1966) assumption that  $u_{st} = u_0$  is no longer valid at high solids flow rate; since it was possible to obtain a moving bed in the downcomer (with no diameter reduction) meanwhile the stages were fluidized.

Knowlton (1986) describes the fundamentals of the overflow standpipes, and analyzes the different bubbling regimes which may be present in the downcomer, depending on the particle Geldart type, and on both the gas and the solid velocities. Knowlton pointed out that there are two main types of regimes inside an overflow standpipe: (a) fixed (or moving) bed flow, when the gas-solids relative velocity,  $u_{slip}$ , is lower than the minimum fluidization velocity  $u_{mf}$ , and (b) fluidized bed flow, when  $u_{slip} > u_{mf}$ . This latter case includes four regimes of fluidized bed flow, depending on the direction of the emulsion and bubble velocities.

Krishnaiah and Varma (1988) developed a simple and effective downcomer for use in gas-solid multistage fluidized beds to facilitate transfer of solids from one stage to the next stage at minimum fluidization velocity. They provided a cone with height to diameter ratio of one, having a discharge opening of 5 mm at the exit end of downcomer. However they did not characterize the regime of flow in the downcomer. For operations at low temperature, these systems can be modified in order to improve the circulation of solids, adding aeration, L-valve, etc. [Knowlton and Hirsan, 1978; Krishnaiah and Verma, 1988], but in operations such as combustors or similar processes at high temperature, the incorporation of these accessories may be extremely difficult. Some researchers [Krishnaiah and Verma, 1982; Krishnaiah and Verma, 1988] included a constriction (cones, cylinders, etc.) at the bottom of the standpipe in order to reduce the gas velocity inside the standpipe, and consequently, widening the stable operating range. Other workers [Rao and Gopalkrishna, 1978] placed an inclined flap below the downcomer exit, in order to reduce the gas flow rate through the downcomers. Zang and Rudolph (1998) studied on flow instability in non-fluidized downcomer/standpipe flow and developed a correlation for design of downcomer for smooth transfer of solids from stage to stage.

#### Distributor

One of the most important problems to be overcome in determining the correct operation of a multistage fluidized-bed reactor is an appropriate choice of a gas distributing element. The choice of distributor and the determination of its basic constructional parameters must take account of a number of criteria (which are quite often contradictory) connected with the hydraulics of processes occurring in the bed together with a simultaneous allowance for the particular features of the operation conditions to be employed. Inadequate design of gas distribution, or their malfunction in operation, is responsible for a substantial proportion of the difficulties encountered in fluidized bed processes. In applications where high conversions of the reacting gases are required, careful design of the distributor so as to give uniform gas distribution and small bubbles at the grid can improve performance. Distributors must also have sufficient strength to resist deformation under operating conditions and to support the static bed. They must be able to withstand stresses induced by thermal expansion, operate long periods without blocking and be easy to unblock, prevent backflow(sifting) of solids into the wind box, avoid promoting erosion of the plate and attrition of particles and operate at as low pressure drop as possible in order to minimize the power consumption. Not all these requirements are compatible and their relative importance may change with the process requirement.

Further, knowledge of gas pressure drop through the distributor  $(\Delta P_d)$  in a fluidized-bed vessel is essential for two basic reasons. Firstly, in connection with an estimation of the total pressure drop in the apparatus, this has a direct influence on the cost of gas transport and an indirect influence on the investment and operational costs of the installation. Secondly, the gas pressure drop through the distributor has a

decisive effect on the stable operation of the fluidized bed. Thus, the system consisting of gas distributor/fluidized bed of solid particles may be considered in terms of the connection between two hydraulic resistances. Any fluidized bed is characterized by the periodic occurrence of local flow non-uniform in both the phases. Such non-uniformity lead to local increases in the gas velocity and a properly designed system of hydraulic resistances should compensate for any perturbations arising from local changes in gas pressure through the distributor.

The state in which all the non-uniformity in the operation of the bed are eliminated, in such a way that a permanent decay in fluidization does not occur in any part of the vessel, is defined as stable fluidization. Numerous experimental studies have shown that this state is linked directly to the ratio  $\Delta P_d / \Delta P_h$  i.e. the ratio of the gas pressure drop through the distributor to the pressure drop across the bed. Qureshi and Creasy (1979) have listed extensive data, which include values for this ratio for various industrial fluidized-bed vessels whose performance has been assumed to be of the correct form. From this it follows that depending on the process type, the construction of the distributor and the kind of fluidized solid employed, values of the ratio lie in the range 0.01-1.0 [Qureshi and Creasy ; 1979]. The ratio  $\Delta P_d / \Delta P_b$ , also has a decisive influence on the number of active holes in the distributor. According to Sathiyamoorthy and Rao (1978), an increase in this ratio leads to an increase of the fraction of active orifices present and hence to a uniform gas distribution over the whole cross-sectional area of the apparatus. In summary, it should be stressed that an increase in the gas pressure drop through a distributing element allows the establishment of a uniform and stable gas distribution in the vessel. However, such a situation also leads to an increase in the operating costs of the system.

Ruzamov et al. (1972) suggested that the gas distributors employed in the vessels of those operating with standpipes are characterized by a free area ratio F < 10%, which ensures that the gas velocity in the orifice exceeds the terminal velocity of the bed particles by a large factor. This, in turn, should prevent the downflow of solids through the holes in the distributor. In the case considered here the solid particles are displaced to a lower stage through a standpipe.

For gas distribution to be even (Kuni and Levenspiel, 1991), the hydrodynamic resistance of the grid should be such that the re-arrangement resistance is of the order of expansion loss for the gas when it passes from inlet connections into the vessel. Overcashier et al., 1959; Agrawal et al. (1962), Whitehead et al. (1967),

Geldart and Baeyens (1985), Baskakov et al.(1985), Sathiyamoorthy and Horio(2003), Carmello et al.(2000), Kuo and Ku(2008) suggested different values of the grid resistance for even distribution of the gas. The grid area which is the sum of the areas of all the openings usually various from 2-5% of the total cross sectional area of the column. For perforated grid plates of several millimeters thickness, Vanecek et al (1966) suggested the following correlation for predicting fractional area which ensures perfect mixing as

$$F = 1.7 \left[ \frac{u_{op}}{u_{mf}} \right]^{0.9} \tag{2.2}$$

Where, F is the free area expressed as the percentage of the total grid or bed area and  $(u_{op}/u_{mf})$  is the ratio of the operating to minimum fluidizing velocity. The diameter of the openings in the grid should be such that clogging by fines is avoided during stoppage. It is recommended that the diameter of the holes should be at least 1/10 of the particles diameter and atmost half the diameter of the largest size particle.

Chyang et al., (2008) studied on effect of distributor design on gas dispersion in a bubbling fluidized bed reactor. For a certain type of distributor, a better extent of gas dispersion can be obtained while distributor with lower opening area ratio is employed.

#### 2.5 Flow properties of solids

The fluidization of solids depends on the properties of solids and fluids. The fluid properties (namely that air) are dependent on temperature and pressure and could be estimated easily or can be found from literature. These properties include air density and viscosity. In case of fluidization of solids; the main properties affecting fluidization are solid particle size, density, porosity and sphericity.

# (a) Particle size (d<sub>p</sub>)

The material particle sizes were determined by standard sieve analysis in B.S. sieves. The only sharp cut fractions were used in the experimentation. The average of the diameters of the sieves through which the solids pass and the sieve on which they are retained is taken as diameter of the particles.

# (b)Density (ρ<sub>s</sub>)

The density of the solids is determined by the usual liquid displacement. To ensure that the wettabillity of the solids by a liquid does not affect the results, the density of lime has been determined by kerosene.

# (c) Porosity (E)

The porosity or void fraction of a solid material of a definite size is determined by knowing the volume of the bed and the volume of the solids. The ratio of the void volume to the volume of the bed gives the porosity of the bed. The porosity of the bed is affected by the diameter of the cylinder in which the volumes are measured. To ensure accuracy in fluidization experiments, the cylinder diameter has been taken same as the diameter of the column.

# (d) Sphericity (Ø<sub>s</sub>)

The sphecicity of the lime particle has been determined by pressure drop measurements. For a given material, the pressure drop is determined in fixed bed zone using air as the medium. In the test carried out the gas velocity was kept in laminar zone by limiting the particles Reymold's number value less than 10. With voidage, pressure drop and other fluid and solid properties known, the sphericity was calculated using Ergun's fixed bed pressure drop equation written in the following form, omitting the second term for kinetic energy losses and substituting  $(d_p\phi_s)$  for  $d_p$  for the case of non-spherical particles.

$$\frac{\Delta P_s}{H} \cdot \frac{d_p^2 \cdot \phi_s^2}{\mu_g \cdot \mu_g} \cdot \frac{\varepsilon_{mf}^3}{(1 - \varepsilon_{mf}^3)} = 150$$
(2.3)

### Minimum Fluidization Velocity, umf

The Minimum fluidizing velocity is one of the important design characteristics of the fluidized beds which explain the transition from fixed bed to fluidized bed condition. The basic principles on which expressions for predicting the minimum fluidizing velocity are based on;

- At the incipient fluidization condition, the bed behaves much the same way as the fixed bed and the bed pressure drop is equal to the buoyant weight per unit cross sectional area.
- (ii) The conditions at the onset of fluidization are similar to that at the free falling condition of a particle and the flow conditions of these two limits are developed.
- (iii) The drag force acting on the particle is equal to the submerged weight of the particles.

Considering the incipient fluidizing condition to be the extreme point in fixed bed conditions, Leva et al.,(1948) attempted a correlation for minimum fluidizing velocity in terms of the system properties, shape factor( $\emptyset_S$ ) and bed voidage ( $\varepsilon_{mf}$ ) at minimum fluidizing conditions. Leva (1956) modified the equation by expressing the unknowns  $\emptyset_S$  and  $\varepsilon_{mf}$  as function of Reynond's number and gave an empirical correlation as

$$G_{mf} = 688d_p^{1.82} \left[ \rho_f (\rho_S - \rho_f) \right]^{0.94} / \mu^{0.88}$$
(2.4)

where  $G_{mf}$  is in pounds per square foot per hour;  $d_p$  is in inches;.  $\rho$  in pounds per cubic foot and  $\mu$  in c.p. Based on large volume of experimental data covering a wide variety of systems, few researchers [Johnson,1950; Wilhelm and Kwauk,1948] developed empirical correlations. Narasimhan (1965), Pinchbeck and Popper (1956), Godard and Richardson (1969), Haider and Levenspiel (1989), Smith (1998) proposed correlations for predicting the minimum fluidizing velocity using the concept of the free falling velocity. Correlations for predicting minimum fluidizing velocity based on drag force considerations have been attempted by Frantz (1966), Pillai and Raja Rao (1971), Balakrishna and Raja Rao (1971), Sengupta and Rao (1971) with reasonable accuracy for a given system. Motamedei and Jameson (1968) attempted to measure minimum fluidizing velocity on the basis of two phase theory of fluidization and concluded that the only safe way to obtain minimum fluidizing velocity is to measure it for individual gas-solid systems.

Ergun (1952) developed an equation giving the pressure drop in a gas flowing thorough a packed bed, such as the so-called Ergun equation:

$$\frac{\Delta P_{s}}{H} = 150 \frac{\left(1 - \varepsilon_{mf}\right)^{2}}{\varepsilon_{mf}^{3}} \frac{\mu_{f} u_{mf}}{d_{p}^{2}} + 1.75 \frac{\left(1 - \varepsilon_{mf}\right)}{\varepsilon_{mf}^{3}} \frac{\rho_{g} u_{mf}}{d_{p}}$$
(2.5)

where the first term represented the viscous energy loss while the second term represented the Kinetic energy loss.

Wen and Yu (1966b) developed an expression for the minimum fluidization velocity for arrange of particle types and sizes by assuming the following approximations to hold based on experimental data:

$$\frac{1-\varepsilon_{mf}}{\phi_s^2 \varepsilon_{mf}^3} \cong 11 \text{ And } (1/\phi_s \varepsilon_{mf}^3) = 14$$
(2.6)

Wen and Yu (1966b) combined these with Ergun equation and obtained the following equation for Reynold's number at minimum fluidization. (which is most widely used),

$$R_{\rm emf} = \sqrt{\left(C_1^2 + C_2 Ar\right) - C_1}$$
(2.7)

They suggested that  $C_1$ = 33.7 and  $C_2$ =0.0408. A large number of values for  $C_1$  and  $C_2$  have been available in the literature.

Recently, there have been several studies on the independent effects of temperature and pressure on  $U_{mf}$ . In fine powders,  $U_{mf}$  decrease with increase in temperature [Svoboda and Hartman, 1981] and is hardly affected by pressure while in coarse powders, increased temperature causes an increase in  $U_{mf}$  and a pressure decrease (King and Harrison, 1982). Svoboda and Hartman (1981) have also reported extensive data on  $U_{mf}$  at high temperature for limestone, lime, coal ash etc. There is good evidence that minimum fluidization velocity is affected by both scale of equipment and distributor design (Gunn and Hilal, 1997).In view of reported literature, it was decided to determine the minimum fluidization velocity of the materials for the given apparatus setting.

#### 2.6 Multistage continuous counter current fluidization

Flow of solids from stage to stage through downcomers depends on the gas flow rate and on the geometry of the apparatus. There is a limited range under which the flow of solids is smooth and beyond which the operation of the column is unsteady. In the present investigations an attempt was made to identify the zone of smooth solids flow along with the hydrodynamic aspects of gas-solid systems. The solids employed in the study, compriseed single systems like hydrated lime, sand (A), sand (B) and the mixed particles like lime-dolomite.

# 2.6.1 Experimental set up

Fig. 2.7 and Fig. 2.8 show the schematic and pictorial representation of the multi-stage fluidized bed reactor designed and developed and used in this study. The configuration of staged gas-solid fluidized bed reactor is similar to that of the sieve trays distillation column. The experimental set-up consisted of three stages fluidization column (FB<sub>1</sub>, FB<sub>2</sub> and FB<sub>3</sub>) having provision of solid feeding from the top and air supplying system from the bottom along with other auxiliary equipments used for experimentation.. Each stage of the column was constructed of perspex cylinder of 0.10 m internal diameter. Since the diameter of the bed has been chosen as 0.1 m, height of the weir has been considered from 0.03 m to 0.07 m to maintain a



Fig. 2. 7. Schematic diagram of the experimental set-up of a three-stage countercurrent fluidized bed reactor



Fig. 2. 8. Pictorial diagram of the experimental set-up of a three stage counter-current fluidized bed reactor

shallow bed which gives a distinct advantage of restricting the bubble size resulting in preventing the formation of slug and then escape without much conversion. The height of downcomer depends on the height of column. The height each stage of the column was chosen as 0.305 m to prevent the contact of the fluidized particles with upper distributor at any point of time or else at any point it choke the orifices of the distributor creating non-uniform fluidization. The stainless steel plates of 0.002 m thick each ( $G_1$ ,  $G_2$  and  $G_3$ ) were used as gas distributor (Fig. 2.5) between two stages.

Each plate was drilled with perforations of 0.002 m diameter on a triangular pitch having 10.56% total grid openings. The perforations were made accurately avoiding possible burrs and protrusions during drilling process. The grid plates were covered with fine wire mesh (100 mesh size) to prevent the flow of solids through the openings. Table 2.1 shows the detail of perforated plate gas distributor;

Charasteristics		Value
Material		Stainless Steel
Thickness		2.0 mm
Orifice size	٥	2.0 mm
Orifice pitch		5.0 mm
Open area		10.56%

Table 2.1. Characteristics of perforated-plate gas distributors

Each section was provided with a downcomer of perspex cylinder of 0.025 m internal diameter ( $D_1$ ,  $D_2$ ,  $D_3$  and  $D_4$ ) and the downcomers were fitted to the gas distributors by special threading arrangement having the provision for adjusting the weir height as desired. The downcomers were further fitted with a cone at the exit end in order to reduce the up flow of the gas through the downcomer and consequently, widening the stable operating range. The bottom-opening diameter of the cone was 0.012 m for flow of solids to the next stage. Pressure tapings were provided just below the grid plate and the near the air out let and four manometers were provided to measure the pressure drop at every stage as well as the total pressure drop. The inner end of the pressure tap was covered by means of 400 wire mesh SS sieve to prevent solids entering into the silicon tube connected to the manometer. Air from a twin-lobe

compressor (E) was used to supply the air as fluidizing gas and its flow rate was measured using a calibrated rotameter (F). A gas distributor of 0.150 m long was provided at the bottom of column for uniform distribution of gas to the column. A conical hopper was attached at the bottom of column for storage of solids coming out from the bottom stage through the downcomer. Fig. 2.8(a) is the pictorial view of outlet storage. The gas leaving the column from the top stage was passed through a 0.15 m diameter standard cyclone (C) and then into the exhaust system. Fig. 2.8(b) is the pictorial view of cyclone used. A cloth bag was attached at the bottom of the cyclone to collect the fines, if any, carried over from the fluidized bed.



Fig. 2.8(a). Photographic view of outlet storage



Fig. 2.8(b). Photographic view of cyclone



Fig. 2. 8(c). Photographic view of feeder

A perspex hopper of 0.150 m internal diameter and 0.4 m long called as feeding funnel was used to hold the lime and attached to the screw feeder .The solids from the screw feeder was fed through a Perspex tube of 0.012 m internal diameter to the first stage downcomer of the reactor. Fig. 2.8(c) is the pictorial view of variable speed screw feeder. The gas flow rate was adjusted by a control valve and measured by a rotameter. Necessary precautions were made to ensure that no air from outside intruded into the column during operation. In the study the particles used were sand, lime and dolomite. The characteristics of bed material used in this study are presented in Table 2.2.

Material	Density,	Particle size,	Minimum bed	Sphericity
	Kg/m <sup>2</sup>	μm	porosity, $\varepsilon_{mf}$	Øs
Lime	2040	426	0.48	0.7
Sand(A)	2650	426	0.48	0.76
Sand(B)		600	0.49	0.76
Dolomite	2700	426	0.50	0.70

Table 2.2. Properties of bed materials

# Experimental determination of minimum fluidization velocity

The minimum fluidization velocity of all investigated materials was determined experimentally in a single stage fluidizer at room temperature and atmospheric pressure. For sake of understanding, the experimental data has been presented in figures for two particles. Figs. 2.9 and 2.10 show the effect of superficial

gas velocity on the bed pressure drop. The minimum fluidization point is defined as the intersection of the extrapolated fixed-bed characteristic with the line of constant bed pressure of the fluidized bed, which gives minimum fluidization velocity.



Fig. 2. 9. Effect of superficial gas velocity  $(u_g)$  on pressure drop due to solids  $(\Delta P_S)$  for sand particles



Fig. 2. 10. Effect of superficial gas velocity  $(u_g)$  on pressure drop due to solids  $(\Delta P_S)$  for lime particles

The comparison of experimental mass velocity of solids at minimum fluidization with values obtained from theoretical equation is presented in Table 2.3.

	T		- mi · mu	
Material	Average	Value of Gmf	Value of Gmf	Experimental
	particle size	based on Leva's	based on Wen	value of Gmf,
1	(μm)	eqn.(kg/m <sup>2</sup> ·s)	and Yu eqn.,	$(kg/m^2 \cdot s)$
			$(kg/m^2 \cdot s)$	
lime	426	0.131	0.121	0.132
Sand(A)	426	0.167	0.142	0.159
Sand(B)	600	0.315	0.274	0.295
Dolomite	426	0.157	0.127	0.142

Table 2.3. Comparison of Gmf value

# 2.7 Studies on pressure drop due to distributor

Because of the major role of the distributor in fluidized bed operation, the problem of calculating the gas pressure drop across a distributor has been the subject of numerous studies. The present investigation was to determine the gas pressure drop across distributor and compare it with the experimental results.

# 2.7.1 Theoretical determination of the gas pressure drop through a distributor

The gas pressure drop through a distributor may generally be considered in a similar manner to that used in the description of local hydraulic resistances. Thus, the general form of the relationship for  $\Delta P_d$ , which results from the Bernoulli equation, may be written as

$$\Delta P_d = \xi \left( \frac{\rho_g u_0^2}{2} \right) \tag{2.8}$$

Since Eq. (2.8) is a transformation of the Body-Carnot equation, it introduces the dimensionless coefficient  $\xi$  which involves the ratio of the energy loss to the kinetic energy of the stream as a means of characterizing the pressure loss. In applying this relationship to the description of the pressure drop through a gas distributing element in a fluidized-bed vessel, various authors have described the coefficient  $\xi$  in different terms.

An analysis of these equations allows their subdivision according to the manner in which they describe this coefficient into:

(i) Kuni and Levenspiel (1991 which described  $\xi$  exclusively as a function of the parameters related to gas flow through a fluidized-bed vessel, i.e.  $\xi = f(R_e)$ .

(ii) Idelchik (1975) which presented  $\xi$  exclusively as a function of the constructional parameters of the distributor, i.e.  $\xi = f(F, d_{or}, p, t)$ .

(iii) Pillay and Varma(1983) which described  $\xi$  as a function of both the flow parameters and of the constructional characteristics of the distributor, i.e.  $\xi = f(\text{Re}, \emptyset_s, d_{\text{or}}, p, t)$ .

The most complete description of the drop in gas pressure through an orifice distributor is given by the group (iii) above. In practice, however, this is at the cost of the complexity of the relationship with the result that the eligibility of these equations is reduced and their application procedure is difficult. On the other hand, the simplicity of formulae belonging to groups (i) and (ii) and their fully satisfactory accuracy for design purposes makes their general application justifiable.

Considering the value of  $\xi = 1/(F.C_d)^2$  described by Kuni and Levenspiel (1991), Eq. (2.8) is modified as follows;

$$\Delta P_d = \frac{G_f^2}{2\rho_g C_d^2 F^2} \tag{2.9}$$

When dealing with a multi-orifice distributor it is usually convenient to express the above equation in terms of pitch/hole diameter.

For triangular pitch 
$$N = \frac{\pi}{2\sqrt{3}} \left(\frac{d_{or}}{p}\right)^2$$
 (2.10)

$$\Delta P_d = \frac{6G_f^2}{\pi^2 C_d^2 \rho_g} \left(\frac{p}{d_{or}}\right)^4 \tag{2.11}$$

The coefficient of discharge as proposed by researchers [Pillay and Verma, 1983] is satisfactorily correlated for range of experimental conditions as given below;  $C_d=0.6974 (u_g)^{0.2}$  (2.12)

Putting the value of  $C_d$  in Eq. (2.11), it gives the theoretical pressure drop through a distributor.

# 2.7.2 Experimental procedure

This section reports on the studies on the pressure drop due to empty column in the multi-stage fluidized bed reactor. The schematic diagram of the experimental set-up is discussed in earlier section of this chapter and is shown in Fig. 2.7. . The pressure taps at each stage of the reactor and whole column were provided and connected to U-tube manometers of 1.0 m long by silicon tubes in order to measure the differential pressure due to distributor and wall at each stage and total pressure drop across the whole column. Carbon tetrachloride (density-1590 kg / m<sup>3</sup>) was used as the manometric fluid. The pressure differences ( $\Delta Ps$ ) for each gas-solids rate at each stage and whole column were measured by the difference in the head ( $\Delta h$ ) of the carbon tetrachloride (CCl<sub>4</sub>) in the limbs of the U-tube manometers. The pressure drop due to distributor and wall can be experimentally calculated from Eq. (2.11).

$$\Delta \mathbf{P} = \Delta \mathbf{h} \left( \rho_l - \rho_g \right) \mathbf{g} \tag{2.13}$$

Where,  $\Delta P$  is differential pressure drop across the section (N/m<sup>2</sup>),  $\Delta h$  the difference in the head of the carbon tetrachloride (CCI<sub>4</sub>) in the U-tube manometer,  $\rho_l$  and  $\rho_g$  are density of the CCI<sub>4</sub> liquid (kg/m<sup>3</sup>) and gas (air) (kg/m<sup>3</sup>) and g is the acceleration due to gravity (m·s<sup>-2</sup>).

Air at room temperature was used as the fluidizing medium. The actual density and viscosity of air at specific fluidization temperature are given below [Wu and Bayens, 1991].

$$\rho_a = 1.2 \, \frac{293}{T} \tag{2.14}$$

$$\mu_a = 1.46 \times 10^{-6} \frac{T^{1.504}}{T + 120} \tag{2.15}$$

### 2.7.3 Results and discussion

The gas pressure drop due to distributor and wall in the absence of the solids were measured at different gas flow rates .No discernible difference in pressure drop due to distributor and wall across each stage was noticed. In view of identical performance, the gas pressure drop due to distributor and wall across each stage were obtained and the variations of pressure drop across a single stage under various flow conditions were presented in Fig. 2.11.The pressure drop increased linearly with gas velocity because distributor offered more resistance with increment in gas velocity [Chyang and Hwang, 1991].

It may be seen in the Fig. 2.12 that the experimental value is in close agreement with theoretical value as predicted using equation (2.12). However, the reason of slightly higher value of experimental pressure drop may be due to screen, which has not been considered in the theoretical equation.



Fig. 2. 11. Effect of superficial mass velocity of gas on pressure drop due to distributor  $(\Delta P_d)$ 



Superficial mass velocity of  $gas(G_x X 10^2)$ , kg/m<sup>2</sup>·s

Fig. 2. 12. Comparison of experimental and predicted effect of superficial mass velocity of gas on pressure drop due to distributor  $(\Delta P_d)$ 

#### 2.8 Summary of the findings

The distributor and downcomer play a major role for stable operation of a multistage fluidized bed reactor. Various design aspects of downcomer and distributor are discussed in the chapter. The minimum fluidization velocity of materials are determined experimentally and compared with theoretical values. The experimental minimum fluidization mass velocities of gas for lime, sand(A) and sand (B) are found to be 0.132, 0.159 and 0.295 kg/m<sup>2</sup> s. The gas pressure drop due to distributor alone has been determined experimentally and compared with theoretical value. It is observed that the gas pressure drop due to distributor is almost same across each stage and increases with increase in gas flow rate with the given apparatus. The reason of slightly higher value of experimental pressure drop compared to theoretical may be due to screen. With the basic data generated in this chapter, the hydrodynamic aspect of the multistage fluidized bed reactor with flow of solids may now be investigated.

# **CHAPTER 3**

# HYDRODYNAMICS OF SINGLE PARTICLE SYSTEM IN A MULTISTAGE FLUIDIZED BED REACTOR
# Hydrodynamics of single particle system in a multistage fluidized bed reactor

#### **3.0 Introduction**

In the present investigation, a counter-current multistage fluidized bed reactor (MFBR) operating in three stages has been designed, constructed and investigated so that the hydrodynamic as well as the mass transfer characteristics can be improved without substantially increasing energy dissipation. In Chapter-2 it has been discussed that multistage fluidized bed reactors, using horizontal perforated plates to segment the gas-solid fluidization column into a number of stages, has gained importance for conducting unit operations such as drying, adsorption, calcinations, regeneration of catalysts etc.. Before using the reactor for specific purpose, the hydrodynamics of the multi-stage fluidized bed reactor must be studied in detail so as to maximize the efficiency of the system. In the present chapter, an attempt has therefore been made to acquire precise knowledge of the hydrodynamic characteristics of MFBR under bubbling regime for two-phase flow in the system.

#### 3.1 Status of research on hydrodynamics of multistage fluidized bed reactor

Theoretically, bubbling fluidized bed reactors may be employed for any gasliquid or gas-solid contacting and mixing process (Askina and Kunreuther, 1951; Van Deemter, 1960]. Practical application however, depends on their ability to achieve the required rates of momentum, heat and mass transfer at acceptable capital and operating costs. The best way to reduce the operating costs of existing units is to improve their efficiency and operation via process optimization and control. To achieve this improvement, a thorough understanding of fluidization principles and how fluidized-bed systems are designed is essential. Haipel (1958) has studied on multi-particle dynamics in single stage fluidized bed reactor indicating slow motion of fluid relative to beds of spherical particles increasing gas residence time in the bed. The gas-fluidization dynamics in single stage bubbling fluidized bed have been studied by Werther (1974), Levy et al. (1983), Lim et al. (1995), Jackson (2000), Ajber et al. (2002), Huilin et al. (2003), Patil et al. (2005), and Dahl et al. (2005). The simplicity of design and construction, better defined flow patterns and comparatively low power inputs for requisite transport rates make the system very attractive for industrial application. However, the amount of generalized information available on hydrodynamics of MFBR and its performance analysis is very few. This is due to the fact that, for a given superficial gas velocity, a large number of variables like gas or solid physical properties, downcomer cross section ratio, solid rate, gas distributing plate etc. affect significantly the performance of the reactor.

Few researchers [Kannan et al, 1994; Kishnaiah and Verma, 1982; Papadatos et al., 1975; Rao and Gopalkrishna, 1972; Nguyen et al., 1973; Pillay and Verma, 1983; Rao and Gopalkrishna, 1978; Martin-Gullon et al., 1995; Verma, 1975; Hymore and Laguerie, 1983; Eleftheriades and Judd, 1978; Lin et al., 1986; Martin-Gullon et al., 1996; Kersten et al., 2003] have investigated on some aspects of multistage fluidized bed reactors with and without downcomer and occasionally confused technological information is encountered in the literature. In view of scanty literature and technical information, studies in multistage continuous counter current fluidization with downcomer have been carried out to predict the hydrodynamic behavior for single particle system.

Fluidization of particles under bubbling regime is possible with gas velocities ranging from minimum fluidization velocity to terminal velocity of the particles. But a counter-current MFBR is susceptible to flooding during its operation [Verma, 1975]. There is a limited range of operation under which the flow of solids is smooth and beyond which the operation of the column is unsteady. The immediate experimental investigations are to identify the mechanism of solids flow and zone of smooth flow of solids in the system.

## 3.2 Experimental set-up and techniques

In the investigation a multistage fluidized bed reactor with downcomer, operating in three stages was designed. Fig .3.1 shows the schematic diagram of the experimental set-up. The concept and operating principle of the reactor has been discussed earlier. The experimental reactor was a three stage vertical column and each section was separated by a gas distributor. Each section was provided with a downcomer and overflow weir. The downcomers were fitted to the gas distributor by special threading arrangement having the provision for adjusting the weir height as desired and fitted with a cone at the exit to facilitate of the column without priming and to arrest the up flow of the gas through the downcomer. The solid feeding was



Fig. 3. 1. Schematic diagram of the experimental set-up of a three-stage countercurrent fluidized bed reactor

done through a variable-speed screw feeder in order to adjust the desired input flow corresponding to each test. The solids outlet from the column were provided from the bottom most downcomer into a Perspex storage vessel. Air was introduced into the column at the bottom and the air exit is provided at the top of the stage. The grid plate was covered with a fine wire mesh (100 mesh size) to support the solids. The system was so arranged that the solid was fed to the first stage by screw feeder through the downcomer. The air from the compressor was properly metered and introduced at the bottom of the column.

At low gas rate, the solids were allowed to fall into the top stage through feeding down comer  $(D_1)$  and solids piled-up over the distributor plate of the top stage just below the downcomer. As the gas rate was increased in small increments, the solids dispersed and started distributing in the top stage under fluidized condition. Further, increments in mass velocity of gas, solids were then transferred from the stage to stage fluidizing in each stage. Then solids continued to flow out into the solid outlet storage from bottom column. The operation of the column was usually smooth and steady when all the stages had sufficient material. The start up was usually difficult. The materials studied included lime, sand (A) and sand (B).

#### 3.3 Mechanism of gas solid contacting in multistage fluidization

As observed, in multi-stage fluidizer with downcomer, the solids down-flow pattern was complicated. Fig. 3.2 shows the solids flow mechanism from one stage to another stage. It was observed that the solids transport was like moving bed in the downcomer. At the end of the downcomer, the solid was discharged into the solid bed under fluidized condition. Since the extreme end of the downcomer was sunk in the bed, the discharge of solids was probably complicated due to presence of other solids and due to change in the direction of flow of solids. Once in the bed, the solids moved across the tray towards the over flow line. The solid movement across the tray was expected to be governed by the number of inter-particle collisions and the air cushion provided by the up-moving gas. At the over flow, the solids moved down to the lower stage through the downcomer. The process was repeated on each stage till the solids were discharged from the column. The solid flow was downward in the down comer, while it was cross flow on the grid and up flow before its entry into overflow weir.



# Mechanism of solids flow 1. Moving bed flow through downcomer 2. Discharge from bottom of downcomer through cone 3. Cross flow across the bed 4. Overflow of solids

Fig. 3. 2. Schematic of solids flow from upper stage to next stage

As observed, the overall flow rate of solids at any mass velocity of gas was controlled by the flow of solids from downcomer to bed, which was considered as a rate-controlling step.

In the entire column; solid flow was counter-current to contaminated gas stream flow. Zenz and Othmer (1960) has proposed a phase diagrams and discussed the case of counter current flow of solids in an up-moving gas stream. Thus the solids flow from feed point to discharge point in a multistage fluidizer can be divided into four basic steps. These can be listed as;

- i) Flow of the solids from screw feeder into the feeding down comer by gravity.
- ii) Discharge of solids from downcomer into the bed.
- iii) Cross flow of solids from downcomer bottom to the over flow point.
- iv) Over flow of solids from the bed into the downcomer of the next stage.

The first two steps were essentially gravity flow of solids where practically no air flow occurred. At the discharge point of the downcomer, parameter affecting the solids flow was the rate of removal of solids from discharge point. The cross flow of solids was functions of the inter particle collisions and the balance of the gravity force and upward thrust on the particles and the total cross flow path. The last step i.e., the overflow of solids into the transfer line connecting the stage below was perhaps governed by the overflow height, gas and solid flow rate besides the diameter of the overflow line.

## 3.4 Study of stable operation

The mass velocity of gas, at which the solids began to spread at top stage and transferred from stage to stage to give equal pressure drop in all the stages of the column, was termed as the lower stable operating velocity or critical gas velocity,  $u_{cr}$ . With an increase in gas velocity beyond the critical gas velocity, the pressure drop across the stage decreased. With a still further increase in the gas rate, the height of the solids in the downcomers reached a minimum value and, at a particular gas rate, there was no transfer of solids from downcomer to bed with time indicating flooding of the upper multistage fluidized bed. The gas velocity at which flooding occurred, was termed as the upper stable operating gas velocity or flooding gas velocity,  $u_{f}$  [Kannan et al., 1994]. Further increment in mass velocity of gas made the system unstable and the solids from the lower bed got entrained through the downcomer and the lower bed got emptied.

## 3.4.1 Results and discussion

# Effect of mass velocity of solids on mass velocity of gas

Figs. 3.3 to 3.5 present the effect of mass velocity of solids on the mass velocity of gas for the different bed materials. The figures indicated the smooth solid flow region for different gas velocities for the given apparatus setting.

From Fig. 3.3 it was observed that a minimum air flow rate of  $21.2 \times 10^{-2}$  kg/m<sup>2</sup> ·s and  $23.0 \times 10^{-2}$  kg/m<sup>2</sup> ·s was required for smooth flow to start for same size particles such as lime and sand (A). Similar trend was also observed for sand (B). This minimum initiation gas velocity was higher than the minimum fluidizing velocity for a particular material. Thus it can be concluded that for smooth solids flow to occur the gas velocity should be greater than G<sub>mf</sub>. As observed from Fig.3.3 to 3.5, solids flow initiated at (G<sub>a</sub>/G<sub>mf</sub>) in the range of 1.5-2.0. The values of (G<sub>a</sub>/G<sub>mf</sub>) for solid flow initiated in continuous multistage fluidizers at a velocity more than minimum fluidization velocity.



Fig. 3. 3. Effect of superficial mass velocity of solids on mass velocity of gas showing stable operating region in multistage fluidized bed for lime particles (426 µm)



Fig. 3. 4. Effect of superficial mass velocity of solids on mass velocity of gas showing stable operating region in multistage fluidized bed for sand(A) particles (426 µm)



Fig. 3. 5. Effect of superficial mass velocity of solids on mass velocity of gas showing stable operating region in multistage fluidized bed for sand (B) particles (600  $\mu$ m)

As observed from Figs. 3.6 and 3.7, the critical gas velocity increased with increase in particle size and solid flow rate. The flooding gas velocity increased with increase in the particle size and decreased with increase in solid flow rate. The critical and flooding gas velocity increased with in increase in particle density. At a particular gas velocity, increasing the solid flow rate increased the gas resistance in the bed resulting in no-fluidization. At a particular solid flow rate, the increment in mass velocity of gas after certain point did not allow the material to flow from downcomer to bed indicating instability in solids flow and at that time, the solids height in the downcomer increased flooding the upper bed. Further increment in mass velocity of gas, the solids entrained from the lower bed, as the downcomer could not withstand the pressure drop. This shows that the range of stable operation of the multistage fluidized bed reactor with downcomers decreases with increase in solids rate, an observation in agreement with Kannan et al. (1994).



Fig. 3. 6. Effect of particle density on stable operating region in multistage fluidized bed for 426 µm particle size



Fig. 3. 7. Effect of particle size on stable operating region in multistage fluidized bed for same particle densities

#### 3.5 Studies on pressure drop

Once the range of stable operating range for the given apparatus was determined, the pressure drop studies of the reactor were conducted within stable operating range for two phase system. The pressure taps at each stage of the reactor and whole column were provided and connected to U-tube manometers by silicon tubes in order to measure the differential pressure at each stage and total pressure drop across the entire column. The pressure differences ( $\Delta Ps$ ) for each gas-solids rate at each stage and entire column were measured by the difference in the head ( $\Delta h$ ) of the carbon tetrachloride (CCl<sub>4</sub>) in the limbs of the U-tube manometers. The pressure drop is experimentally calculated using Eq. (3.1).

$$\Delta \mathbf{P} = \Delta \mathbf{h} \left( \rho_{\rm L} - \rho_{\rm G} \right) \mathbf{g} \tag{3.1}$$

Where,  $\Delta P$  is differential pressure drop across the section (N/m<sup>2</sup>),  $\Delta h$  the difference in the head of the CCI<sub>4</sub> in the U-tube manometer,  $\rho_L$  and  $\rho_G$  are density of the CCI<sub>4</sub> liquid (kg/m<sup>3</sup>) and gas (air) (kg/m<sup>3</sup>) and g is the acceleration due to gravity (m·s<sup>-2</sup>). The operating experimental conditions are given in Table 3.1.

Parameters	Values
Ambient Temperature (K)	$308 \pm 3$
Mass velocity of gas $(kg/m^2 \cdot s)$	31.27 x 10 <sup>-2</sup> -56.40 x 10 <sup>-2</sup>
Mass velocity of solids $(kg/m^2 \cdot s)$	$35.4 \ge 10^{-3} - 141.5 \ge 10^{-3}$
$u_{op}/u_{mf}$	2.3 – 4.3
Weir height (m)	0.03 - 0.07

Table 3.1. The operating range of variables and experimental conditions

The gas pressure drop corresponding to perforated plate in absence of solids was measured at different mass velocity of gas and the variation of pressure drop with gas velocity discussed earlier as shown in Fig. 2.11. While operating the system with solids, the system was considered to be stable when it had been running evenly for at least half an hour. Under stable condition, it was observed that all the stages of the reactor were identical in their operation as well as performance. The pressure drops across each stage and across the entire column were recorded. No discernible difference in the pressure drop across the stage was noticed from stage to stage. In view of identical performance, the pressure drop due to solids was obtained from the difference between the total column pressure drop with and without solids. Dividing it by the number of the stages gives  $\Delta Ps$ , the pressure drop due to solids per stage of the multistage fluidization column.

$$\Delta P_{T} = \Delta P_{p} + \Delta P_{w} + \Delta P_{s}$$

$$\Delta P_{s} = \Delta P_{T} - (\Delta P_{p} + \Delta P_{w})$$
(3.2)
(3.3)

#### 3.5.1 Results and discussion

# Effect of superficial mass velocity of gas on pressure drop for lime particles

Figs. 3.8 to 3.10 describe the effect of mass velocity of gas on pressure-drop due to lime particles across each stage at different mass velocity of solids and weir heights. It may be seen from the figures that the pressure drop due to solids,  $\Delta Ps$ , decreased with increase in the mass velocity of gas. This was mainly due to the fact that at higher mass velocity of gas, the movement of the particles increases in bed leading to outflow resulting in decrease in solids concentration in bed. The decrease in solids concentration decreases the frictional and impact forces between gas-solid resulting in decrease in pressure drop [Mohanty and Meikap, 2008d]. Similar observations were reported by Pillay and Varma (1983), Krishnaiah and Verma (1982), Kannan et al. (1994). It was observed that the minimum pressure drop occurred at minimum mass velocity of solids corresponding to maximum mass velocity of gas and the maximum pressure drop occurred at minimum mass velocity of gas corresponding to maximum mass velocity of solids. The minimum pressure drops occurred in the column at high mass velocity of gas (56.4 x  $10^{-2}$  kg/m<sup>2</sup>·s) corresponding to minimum mass velocity of solids (35.4 x  $10^{-3}$  kg/m<sup>2</sup> ·s) is 57.0, 103.3 and 143.1  $N/m^2$  at 0.03, 0.05 and 0.07 m weir height respectively. The maximum pressure drops occurred in the column at low mass velocity of gas (31.2 x  $10^{-2}$  kg/m<sup>2</sup>·s) corresponding to maximum mass velocity of solids (141.5 x  $10^{-3}$ kg/m<sup>2</sup>·s) are 98.4, 139.6 and 185.1 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. At a particular mass velocity of solids and gas, increasing the weir height increases the bed volume resulting in increase in solids concentration and thus, pressure drop. The total column pressure drop was found to be in the range of 170 to 555 Pa for the given operating condition.



Fig. 3. 8. Effect of superficial mass velocity of gas (G<sub>a</sub>) on pressure drop ( $\Delta P_s$ ) for lime particles at  $h_W = 0.03$  m



Fig. 3. 9. Effect of superficial mass velocity of gas (G<sub>a</sub>) on pressure drop ( $\Delta P_s$ ) for lime particles at  $h_W = 0.05$  m



Fig. 3. 10. Effect of superficial mass velocity of gas (G<sub>a</sub>) on pressure drop ( $\Delta P_s$ ) for lime particles at  $h_W = 0.07$  m

#### Effect of superficial mass velocity of gas on pressure drop for sand particles

Figs. 3.11 to 3.13 describe the pressure-drop for sand (A) of 426 micron diameter particles measured across each stage varying the gas, solid flow rates and weir heights following the same procedure as described in case of lime particles. Similar trends like lime particles were observed. This may be due to the frictional losses experienced by the gas while traveling through the bed. Although the size of sand and lime particles are same, the pressure drop due to sand particles is more compared to lime particles due to more frictional losses as the density of sand is more than lime particles. The minimum pressure drops occurred in the column at high mass velocity of gas (56.4 x  $10^{-2}$  kg/m<sup>2</sup>·s) corresponding to minimum solid flow rate (35.4 x  $10^{-3}$  kg/m<sup>2</sup> ·s) are 106.4, 159 and 209.5 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. The maximum pressure drops occurred in the column at low mass velocity of gas (31.2 x  $10^{-2}$  kg/m<sup>2</sup>·s) corresponding to maximum solid flow rate (141.5 x 10<sup>-3</sup> kg/m<sup>2</sup>·s) are 185, 257.2 and 364.1 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. It is emerged that at a particular weir height, a decrease in mass velocity of gas decreases the porosity of the bed in the system resulting in increase in solids concentration and hence, pressure drop across the stage [Mohanty and Meikap, 2008c]. At a particular gas and solid flow rate, decreasing the weir height decreases the bed volume resulting in decrease in solids concentration and thus, pressure drop. The total column pressure drop is found to be in the range of 318 to 1092 Pa.



Fig. 3. 11. Effect of m superficial mass velocity of gas (G<sub>a</sub>) on pressure drop ( $\Delta P_S$ ) due to sand (A) particles at  $h_W = 0.03$  m



Fig. 3. 12. Effect of superficial mass velocity of gas (G<sub>a</sub>) on pressure drop ( $\Delta P_S$ ) for sand (A) particles at  $h_W = 0.05$  m



Fig. 3. 13. Effect of superficial mass velocity of gas (G<sub>a</sub>) on pressure drop ( $\Delta P_S$ ) for sand (A) particles at  $h_W = 0.07$  m

#### Effect of superficial mass velocity of gas on pressure drop for sand (B) particles

Fig. 3.14 describes the pressure-drop due to sand (B) measured across each stage varying the gas, solid flow rates at a particular weir height. Similar trend was also emerged for sand particles. The minimum pressure drop occurred in the column at high mass velocity of gas  $(75 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s})$  corresponding to minimum solid flow rate (35.4 x  $10^{-3} \text{ kg/m}^2 \cdot \text{s})$  is 184.0 N/m<sup>2</sup> at 0.05 m weir height. The maximum pressure drop occurred in the column at low mass velocity of gas (56.4 x  $10^{-2} \text{ kg/m}^2 \cdot \text{s})$  corresponding to maximum solid flow rate (141.5 x  $10^{-3} \text{ kg/m}^2 \cdot \text{s})$  is 253.2 N/m<sup>2</sup> at 0.05 m weir height. The results of the experiment indicate that at a particular weir height, an increase in mass velocity of gas decreases the solids concentration resulting in decrease in frictional losses and hence, pressure drop across the stage. The total column pressure drop is found to be in the range of 552 to 759 Pa at the weir height.



Fig. 3. 14. Effect of superficial mass velocity of gas (Ga) on pressure drop ( $\Delta P_s$ ) due to sand(B) particles at  $h_W = 0.05$  m

#### Effect of superficial mass velocity of solids on pressure drop for lime particles

Figs. 3.15 to 3.17 describe the pressure-drop due to lime particles measured across each stage varying the mass velocity of solids and weir heights. It may be seen from the figures that the pressure drop due to solids,  $\Delta Ps$ , increased with increase in the mass velocity of solids. It indicates that higher mass velocity of solids offers a greater volume of solids, which increases the gas-solid interaction. The rise in pressure drop is primarily due to increased particle-wall, particle-gas interaction collectively called frictional losses. This interaction might led to the development of strong aerodynamic ,hydrodynamic and other disruptive forces within gas-solid system at the expense of the gas motive pressure in form of pressure losses or pressure drop. The particle-wall interaction is considered as very small. The minimum pressure drops occurred in the column at high solid flow rate (141.5 x  $10^{-3}$  kg/m<sup>2</sup>·s) are 73.0, 115.1 and 158.2 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. The maximum pressure drops occurred in the column at minimum solid flow rate (35.4 x  $10^{-3}$  kg/m<sup>2</sup> ·s) are 80.0, 126.0 and 170.3 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. The maximum energy lost in form of total maximum column pressure drop at minimum solids flow rate was found to be in the range of 240 to 510 Pa and minimum column pressure drop at maximum solids flow rate 219 to 474 Pa. This was in well agreement with the observations as reported by Pillay and Varma (1983), Krishnaiah and Verma (1982), Kannan et al. (1994). The value of total pressure drop was quite low compared to other conventional wet devices available in market today.



Fig. 3. 15. Effect of superficial mass velocity of solids (G<sub>S</sub>) on pressure drop ( $\Delta P_S$ ) due to lime particles at  $h_W = 0.03$  m



Fig. 3. 16. Effect of superficial mass velocity of solids (G<sub>s</sub>) on pressure drop ( $\Delta P_s$ ) for lime particles at  $h_W$ = 0.05 m



Fig. 3. 17. Effect of superficial mass velocity of solids (G<sub>S</sub>) on pressure drop ( $\Delta P_S$ ) for lime particles at  $h_W = 0.07$  m

#### Effect of superficial mass velocity of solids on pressure drop for sand (A) particles

Figs. 3.18 to 3.20 describe the pressure-drop due to sand (A) particles measured across each stage varying the mass velocity of solids and weir heights. Similar trend was also observed. With increase in mass velocity of solids, particleparticle spacing decreases; as a result the frictional pressure drop increases. At a particular mass velocity of gas and solid, increasing the weir height increases the bed volume resulting in increase in particles concentration and thus, pressure drop. Similar observations were reported by Pillay and Varma (1983), Krishnaiah and The minimum pressure drops occurred in the Verma (1982), Kannan et al. (1994). column at high solid flow rate (141.5 x  $10^{-3}$  kg/m<sup>2</sup>·s) were 148.5, 206 and 291.2 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. The maximum pressure drops occurred in the column at minimum solid flow rate (35.4 x  $10^{-3}$  kg/m<sup>2</sup> ·s) were 132.5, 199.1 and 260.3  $N/m^2$  at 0.03, 0.05 and 0.07 m weir height respectively. The total maximum column pressure drop at minimum solids flow rate was found to be in the range of 396 to 780 Pa and minimum column pressure drop at maximum solids flow rate 444 to 873 Pa.



Fig. 3. 18. Effect of superficial mass velocity of solids (G<sub>S</sub>) on pressure drop ( $\Delta P_S$ ) for sand (A) particles at  $h_W = 0.03$  m



Fig. 3. 19. Effect of superficial mass velocity of solids (G<sub>S</sub>) on pressure drop ( $\Delta P_S$ ) for sand (A) particles at  $h_W = 0.05$  m



Fig. 3. 20. Effect of superficial mass velocity of solids (G<sub>S</sub>) on pressure drop ( $\Delta P_S$ ) for sand (A) particles at  $h_W = 0.07$  m

#### 3.5.2 Theoretical determination of friction factor

Theoretically, the equation which gives the friction factor is as follows [Mohanty and Meikap, 2008d];

$$f = \frac{\Delta p_s}{0.5\rho_g u_g^2} \cdot \frac{d_p}{h_w}$$
(3.4)

#### **3.5.3 Empirical correlation**

An attempt has been made to correlate the friction factor with variables of the system. Conceivable variables including the variables in Eq. (3.4) on which the friction factor in the present system may depend are:

(a) flow properties- gas velocity  $(u_g)$  and solids velocity  $(u_s)$ ;

(b) geometrical properties – height of the weir  $(h_w)$ , and diameter of particles(dp);

(c) physical properties—namely the density of gas ( $\rho_g$ ), density of solid ( $\rho_s$ ) and gravitational constant (g).

The friction factor thus becomes a function of following sensitive parameters, each of them trying to exert it influences:  $f = f[u_s, u_g, g, d_p, h_w, \rho_s, \rho_g]$ 

The variables grouped into dimensionless numbers by employing Buckingham's  $\pi$  theorem, which yields the following equation;

The dimensionless analysis is  $f = k \left[ \frac{u_s}{u_g} \right]^a \left[ \frac{gd_p}{u_g^2} \right]^b \left[ \frac{d_p}{h_w} \right]^c \left[ \frac{\rho_s - \rho_g}{\rho_s} \right]^d$  (3.5)

In order to establish the functional relationship between friction factor and the various dimensional groups in Eq. (3.5), multiple linear regression analysis [Douglas et al., 2001] has been performed to evaluate the constants and coefficients of the equation [Rudolf and Paul, 1979]. The most closely related correlation on the statistical analysis which yields the minimum percentage error, presents the best possible correlation [Mohanty and Meikap, 2008d] as follows:

$$f = 9.0 \left[ \frac{u_s}{u_g} \right]^{0.24} \left[ \frac{gd_p}{u_g^2} \right]^{1.12} \left[ \frac{d_p}{h_w} \right]^{0.2} \left[ \frac{\rho_s - \rho_g}{\rho_s} \right]$$
(3.6)

The correlation coefficient and the standard deviation of the experimental data from regression analysis are found to be 0.8738 and 4.43 respectively. The predicted values of friction factor ( $f_p$ ) from Eq. (3.6) have been plotted against the experimental values ( $f_e$ ) in Fig. 3.21. The comparison between the experimental friction factor and that of predicted from the model indicates that there is an excellent agreement with minimum percentage error. The deviation of the model from the experimental values is found to be within 25 % and it is presented in Fig. 3.22.



Fig. 3. 21. Comparison of the experimental and predicted friction factor in multistage counter-current fluidized bed reactor



Fig. 3. 22. Deviation between experimental and theoretical friction factor in multistage counter-current fluidized bed reactor

#### 3.6 Studies on solid hold-up

The expression 'solid hold-up' is used to mean the amount of solids retained on each plate when the fluidized bed column is in operation. For each experiment, the system was allowed to reach equilibrium so that the inlet and outlet solids flow rate were equal. All the stages of the reactor were identical in their operation as well as performance. The gas and the solids flow was then cutoff simultaneously and the solids were weighed. In view of identical performance and equal solids holdup, the variations of solids holdup across a single stage under various flow conditions have been presented.

#### 3.6.1 Results and discussion

# Effect of superficial mass velocity of solids on holdup (lime particles)

Figs. 3.23 to 3.25 show the effect of mass velocity of solids on solids holdup (lime) measured across a single stage varying the gas and weir heights. It may be seen from the figures that the solids holdup,  $W_e$ , increased with increase in the mass velocity of solids and increased with increase in weir height. The minimum solids

holdup obtained in the column at high mass velocity of gas (56.4 x  $10^{-2}$  kg/m<sup>2</sup> ·s) corresponding to minimum solid flow rate (35.4 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s) were 0.049, 0.09, 0.123 kg at 0.03, 0.05, 0.07 m weir height respectively. The maximum solids holdup obtained in the column at low mass velocity of gas (31.27 x  $10^{-2}$  kg/m<sup>2</sup> ·s) corresponding to maximum solid flow rate (141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s) were 0.084, 0.12 and 0.159 kg at 0.03, 0.05 and 0.07m weir height respectively. The maximum solid holdup at minimum solid flow rate  $(35.4 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s})$  were 0.067, 0.108, 0.146 kg at 0.03, 0.05, 0.07m weir height respectively. The minimum solid holdup at maximum solid flow rate (141.5 x  $10^{-3}$  kg/m<sup>2</sup> ·s) were 0.063, 0.099, 0.136 kg at 0.03, 0.05, 0.07 m weir height respectively. The reason may be that at a particular mass velocity of gas, an increase in mass velocity of solids decreases bed porosity resulting in increase in solids concentration and hence the solids holdup at the stage, as the height of the fluidized bed in the system corresponds to down-comer weir height. Increasing the weir height increases the solids concentration resulting in increasing the solids holdup [Mohanty et al., 2008b]. The range of total solid holdup in the column at maximum solid flow rate varied from 0.190 to 0.480 kg and at maximum mass velocity of gas from 0.147 to 0.401 kg.



Fig. 3. 23. Effect of superficial mass velocity of solids (G<sub>S</sub>) on solids holdup (W) at different weir heights for lime particles and  $G_a = 31.2 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 3. 24. Effect of superficial mass velocity of solids (G<sub>s</sub>) on solids holdup (W) at different weir heights for lime particles and  $G_a = 43.9 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 3. 25. Effect of superficial mass velocity of solids (G<sub>S</sub>) on solids holdup (W) at different weir heights for lime particles (426  $\mu$ m) and G<sub>a</sub>= 56.4 X 10<sup>-2</sup> kg/m<sup>2</sup> ·s

# Effect of superficial mass velocity of solids on holdup sand (A) particles

Figs. 3.26 to 3.28 describe the effect of mass velocity of sand on holdup measured across a single stage varying the gas, solid flow rates and weir heights. It was seen from the figures that the solids holdup,  $W_e$ , increased with increase in the mass velocity of solids and increased with increase in weir height. (Pillay and Varma, 1983; Krishnaiah and Verma, 1982; Mohanty et al., 2008b). The minimum solids holdup obtained in each stage at high mass velocity of gas (56.4 x  $10^{-2}$  kg/m<sup>2</sup> ·s) corresponding to minimum solid flow rate (35.4 x  $10^{-3}$  kg/m<sup>2</sup> ·s) were 0.91, 0.132 and 0.180 kg at 0.03, 0.05 and 0.07 m weir height respectively .The maximum solids holdup obtained in the column at low mass velocity of gas (31.27 x  $10^{-2}$  kg/m<sup>2</sup> ·s) corresponding to maximum solid flow rate (141.5 x  $10^{-3}$  kg/m<sup>2</sup> ·s) were 0.158, 0.221 and 0.314 kg at 0.03, 0.05 and 0.07 m weir height respectively. Increasing the weir height increases solid holding capacity of bed resulting in increasing the solids holdup. The range of total solid holdup in the column at maximum mass velocity of gas varied from 0.273 to 0.75 kg and at minimum mass velocity of gas from 0.339 to 0.912 kg.



Fig. 3. 26. Effect of superficial mass velocity of solids (G<sub>S</sub>) on solids holdup (W) at different weir height for sand (A) particles and  $G_a = 31.3 \times 10^{-2} \text{ kg/m}^2 \text{ s}$ 



Fig. 3. 27. Effect of superficial mass velocity of solids (G<sub>S</sub>) on solids holdup (W) at different weir heights for sand (A) particles and  $G_a = 43.9 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 3. 28. Effect of superficial mass velocity of solids (G<sub>S</sub>) on solids holdup (W) at different weir heights for sand (A) particles and  $G_a = 56.4 \times 10^{-2} \text{ kg/m}^2 \text{ s}$ 

## 3.6.2 Correlations for solid hold-up

Theoretically, the relation between pressure drop and solids hold-up may be written as;

$$\Delta P_{\rm s} = K \, (W/A) \tag{3.7}$$

For ideal condition, K=1.0. In the experiment, K value is found to vary from 0.9 to 0.96, indicating that more than 90% of the material in the stage at any instant is in fluidized state.

It is found experimentally that the solids hold-up is strongly dependent on the pressure drop due to solids in the system. Since a correlation for the pressured drop in terms of the basic parameters of the multi-stage fluidized bed reactor was already been developed, an attempt was made to develop a correlation to find out solids hold-up from operating variables.

Conceivable variables on which the solids hold-up factor in the present system may depend are:

(a) Flow properties- gas velocity (ug) and solids velocity (us);

(b) Geometrical properties –cross-sectional area of reactor (A), height of the weir (h<sub>w</sub>), and diameter of particles (dp);

(c) Physical properties—namely the density of gas  $(\rho_g)$ , density of solid  $(\rho_s)$  and gravitational constant (g).

In order to establish the functional relationship between solids-hold-up and the various dimensional groups, multiple linear regression analysis has been performed to evaluate the constants and coefficients of the equation. It can be seen that the following equation, which yields the minimum percentage error, presents the best possible correlation [Mohanty et al., 2008b]:

$$\frac{W}{Ah_w\rho_s} = 5.0 \left[ \frac{u_s}{u_g} \right]^{0.24} \left[ \frac{gd_p}{u_g^2} \right]^{0.12} \left[ \frac{d_p}{h_w} \right]^{0.2}$$
(3.8)

The coefficient of correlation is of the order of 0.83 and standard deviation of the experimental data from regression analysis is found to be relatively high. To check the consistency of the experimental data, the values of solids hold-up predicted  $(W_p)$ using Eq. (3.8) have been plotted against the experimental values(W<sub>e</sub>) in Fig. 3.29, where the solid line represents the regression equation and points are experimental values. Fig. 3.30 represents the percentage deviation between the experimental data and those predicted by Eq. (3.8) and the percentage deviation to be in the range of  $\pm$  25%. Thus, the empirical correlation satisfies the experimental data of the present system satisfactorily.



Fig. 3. 29. Comparison of experimental values of solids holdup  $(W_e)$  with that predicted from Eq. (3.8)



Fig. 3. 30. Deviation between experimental and predicted solids hold-up

#### 3.7 Mean holding time of solids

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The mean solids holding time,  $\bar{t}$ , is defined as the amount of solids retained in each stage divided by the solid flow rate at stable operating condition. Mathematically, it can be written as;

Solids holdup in the stage (W)

Solids flow rate, m (=AGs)

The above equation is satisfactorily correlated by defining a modified dimensionless holding time,  $\theta$  [Krishnaiah and Verma, 1982].

$$\theta_{e} = \frac{\bar{t}\gamma}{d_{p}h_{w}}$$
(3.10)

$$\theta_{p} = (1 - \varepsilon) \frac{\gamma}{u_{s} d_{p}}$$
(3.11)

#### 3.7.1 Results and discussion

Fig.3.31 describes the comparison between the experimental mean holding time and predicted mean holding time. The mean solids holding time is satisfactorily correlated as shown in Fig.3.31 by defining a modified dimensionless holding time,  $\theta$ . The coefficient of correlation is of the order of 0.98 and standard deviation of the experimental data from regression analysis is found to be marginal. The mean residence time decreased with the increase of the gas velocity. Since increasing in the gas velocity increases the gas holdup in the bed at a particular time, it decreases the residence time of particles in the bed.



Fig. 3. 31. Comparison of experimental values of mean solids holding time ( $\theta e$ ) with that predicted mean solids holding time ( $\theta_P$ )

#### 3.8 Theoretical analysis of downcomer

The pressure drops of the gas circulating upwards through the standpipe and of the gas circulating upwards outside the standpipe are equal [Knowlton, 1986]. Balancing the pressure drop inside and outside the downcomer in absence of solids on the plate, the pressure drop of gas across the plate equals the pressure drop across the downcomer, the latter being the sum of pressure drop due to constriction and due to skin friction (wall) in the downcomer.

Accordingly, 
$$\Delta P_P = \Delta P_c + \Delta P_w$$
 (3.12)

When the system is in operation with solids on the bed, the pressure drop across the downcomer ( $\Delta P_c + \Delta P_w + \Delta P_{dc}$ ) is equal to the pressure drop due to upper bed and upper distributor ( $\Delta P_p + \Delta P_b$ ).

$$\Delta P_{c} + \Delta P_{w} + \Delta P_{dc} = \Delta P_{p} + \Delta P_{b}$$
(3.13)

The pressure drop due to distributor or perforated plate is given by Eq. (2.11). The minimum condition to be satisfied for stable operation is that pressure drop caused by solids inside downcomer must be greater than pressure drop due to upper bed and upper distributor (neglecting the pressure drop due to constriction and wall inside the downcomer). Then the Eq. (3.13) should be;

$$\Delta P_{dc} > \Delta P_p + \Delta P_b \tag{3.14}$$

$$\Delta P_{dc} = K_1 (\Delta P_p + \Delta P_b) \tag{3.15}$$

The pressure drop to be supported in the downcomer due to moving solid particles with porosity corresponding to the minimum fluidization velocity may be taken as

$$\Delta P_{dc} = \rho_s g h_s (1 - \varepsilon_{mf})$$
(3.16)

The theoretical equation, which gives the friction factor as follows;

$$f = \frac{\Delta p_b}{0.5\rho_g u_g^2} \cdot \frac{d_p}{h_w}$$
(3.17)

Putting the value of  $\Delta P_b$  and  $\Delta P_p$  in the Eq. (3.15) and simplifying, we get that the solid height in the downcomer is as follows;

$$h_{s} = \frac{K_{1}\rho_{g}u_{g}^{2}}{2\rho_{s}(1-\varepsilon_{mf})g} \left[\frac{fh_{w}}{d_{p}} + \frac{1}{(FC_{d})^{2}}\right]$$
(3.18)

The solid height in the downcomer is experimentally measured and observed that proper solid height has to be maintained inside the downcomer for stable transfer of solids from upper stage to lower one. If the required solid height in the downcomer is not maintained, the system is unstable.

Analyzing the experimental data, it is found that the value of  $K_1$  comes in the range 4.8 to 5.0. Taking a reasonable value and putting the value in Eq. (3.18) and simplifying, it becomes;

$$h_{s} = \frac{\rho_{g} u_{g}^{2}}{4\rho_{s}(1 - \varepsilon_{mf})} \left[ \frac{fh_{w}}{d_{p}} + \frac{1}{(FC_{d})^{2}} \right]$$
(3.19)

## 3.8.1 Results and discussion

# Effect of mass velocity of solids on downcomer solids height for lime particles

The solid height in the downcomer was experimentally measured and found to increase with increase in the mass velocity of solids and decrease with increase in mass velocity of gas. Figs. 3.32 to 3.34 represent the effect of mass velocity of solids on solids height in the downcomer for lime particles. It was observed that for a mass velocity of gas, proper solid height had to be there inside the downcomer for stable transfer of solids from upper stage to lower one. If the required solid height in the downcomer was not sustained, the system was unsteady. The maximum and minimum solids height was observed to be 0.045m and 0.12 m respectively for lime particles for the given operating conditions.



Fig. 3. 32. Effect of superficial mass velocity of solids on solids height in the downcomer for lime particles at  $h_W$  =0.03 m



Fig. 3. 33. Effect of superficial mass velocity of solids on solids height in the downcomer for lime particles at  $h_W = 0.05$  m



Fig. 3. 34. Effect of superficial mass velocity of solids on solids height in the downcomer for lime particles at  $h_W$  =0.07 m

# Effect of superficial mass velocity of solids on downcomer solids height for sand particles

Figs. 3.35 to 3.37 represent the effect of mass velocity of solids on solids height in the downcomer for sand (A) particles. Similar trend like lime particles was observed in the present case. It was observed that solids height in downcomer increased while increasing the mass velocity of solids and decreased while increasing the mass velocity of gas. The maximum and minimum solids height was observed to be 0.075 m and 0.221 for sand particles for the range of operating conditions. It was observed that height of solids in downcomer was more compared to lime particles. Since the pressure drop due to sand particles is more compared to lime particles at same operating condition, the downcomer has to withstand the more pressure drop resulting in increased solids height in downcomer.



Fig. 3. 35. Effect of superficial mass velocity of solids on solids height in the downcomer for sand (A) particles at  $h_W = 0.03$  m



Fig. 3. 36. Effect of superficial mass velocity of solids on solids height in the downcomer for sand (A) at  $h_W = 0.05 \text{ m}$ 



Fig. 3. 37. Effect of superficial mass velocity of solids on solids height in the downcomer for sand (A) particles at  $h_W = 0.07$  m

# Effect of superficial mass velocity of gas on downcomer solids height

Figs. 3.38 and 3.39 show the effect of mass velocity of gas on solids height in downcomer at 0.05 m weir height for both the materials. It was observed that for a particular solid flow rate, the height of the solids in the downcomers decreased when mass velocity of gas was increased. The experiment indicate that increasing the mass velocity of gas decreases particle concentration in the bed, thereby reducing the total pressure drop due to bed and distributor which is counterbalanced by a reduced solids height in the downcomer. The minimum and maximum solids height in downcomer for the particular weir height was 0.072 and 0.088 m respectively for lime particles and 0.111 and 0.158 m for sand particles.



Fig. 3. 38. Effect of superficial mass velocity of gas (Ga) on solids height in the downcomer  $(h_s)$  for different particles at weir height= 0.05m.



Fig. 3. 39. Effect of superficial mass velocity of gas (Ga) on solids height in the downcomer  $(h_s)$  for different particles at weir height= 0.05m.

#### Comparison of experimental and theoretical solids height

The predicted solids height in the downcomer has been calculated by using Eq. (3.19). Fig. 3.40 shows the comparison of experimental data of present study and predicted solid height in downcomer and suggests that the theoretical solids height derived from Eq. (3.19) is in close agreement with experimental data. The coefficient of correlation is of the order of 0.89 and standard deviation of the experimental data from regression analysis is found to be 2.21. It was observed from the graph that experimental and theoretical solids height in downcomer for lime particles matched very well.

Fig. 3.41 describes the deviation of theoretical solids from the experimental solids height in the downcomer. It may be observed that the percentage deviation is with in the range of  $\pm$  5%.


Fig. 3. 40. Comparison of experimental solids height in the downcomer with that solids height predicated from Eq. (3.19)



Fig. 3. 41. Deviation between experimental and theoretical solids height in downcomer

### 3.9 Studies on aspect ratio

For uniform fluidization (free from channeling),  $\Delta P_d / \Delta P_b$ , (R) is said to fall in range of 0.1-0.4 [Hibey, 1964]. Rice and Wilhelm (1958), Siegel (1976) developed some correlation to select aspect ratio (h<sub>w</sub>/D) corresponding to pressure drop ratio ( $\Delta P_d / \Delta P_b$ ) to maintain a uniform fluidization. An attempt has been made to analyze the relation between aspect ratio and distributor to bed pressure drop ratio with respect to uniform fluidization.

## 3.9.1 Results and discussion

# Effect of aspect ratio on distributor to bed pressure drop ratio for lime particles

Figs. 3.42 to 3.45 present the effect of aspect ratio on distributor to bed pressure drop ratio (R) at a particular solids flow rate corresponding to different operating gas velocity for bed material lime. The distributor to bed pressure drops ratio (R) steeply increased with decrease in aspect ratio revealing its influence on the uniformity of fluidization As the aspect ratio increased, the distributor to bed pressure drops ratio (R) decreased indicating improvement in quality of fluidization and approaching a steady state [Sathiyamoorthy and Horio, 2003] and R increased exponentially with increase in operating velocity for lime particles. In case of lime particles, R value is highest at lowest aspect ratio suggesting agglomeration of lime particles (due to hygroscopic/sticky characteristic) creating dead zones causing non-uniform fluidization.



Fig. 3. 42. Effect of aspect ratio on distributor to bed pressure drop ratio for lime particles and  $G_s = 35.4 \times 10^{-3} \text{ kg/m}^2 \text{ s}$ 



Fig. 3. 43. Effect of aspect ratio on distributor to bed pressure drop ratio for lime particles and  $G_s = 71.0 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 3. 44. Effect of aspect ratio on distributor to bed pressure drop ratio for lime particles and  $G_s = 106.2 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 3. 45. Effect of aspect ratio on distributor to bed pressure drop ratio for lime particles and  $G_s = 141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 

## Effect of aspect ratio on distributor to bed pressure drop ratio for sand particles

Figs. 3.46 to 3.49 represent the effect of aspect ratio on distributor to bed pressure drop ratio (R) at a particular solids flow rate corresponding to different operating gas velocity for bed material sand. The distributor to bed pressure drops ratio (R) increased with decrease in aspect ratio revealing its influence on the uniformity of fluidization. However, the increase in R while decreasing the aspect ratio was not steep as observed in case of lime particles indicating improvement in quality of fluidization and approaching a steady state.



Fig. 3. 46. Effect of aspect ratio on distributor to bed pressure drop ratio for sand (A) particles and  $G_s=35.4 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 3. 47. Effect of aspect ratio on distributor to bed pressure drop ratio for sand (A) particles and  $G_s = 71.0 \times 10^{-3} \text{ kg/m}^2 \text{ s}$ 



Fig. 3. 48. Effect of aspect ratio on distributor to bed pressure drop ratio for sand(A) particles and  $G_s = 106.2 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 3. 49. Effect of aspect ratio on distributor to bed pressure drop ratio for sand(A) particles and  $G_s$ = 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s

## 3.10 Summary of the findings

The formation of fluidized beds in all stages of a multistage fluidized bed reactor is not instantaneous. The stages fill-up in sequences from top to bottom and the approach to steady state is attained in sequence in the system. A minimum solid flow rate is required to form a fluidized bed in each stage in the system. There is a limited range of stable operation in MFBR with downcomer.

- 1. The hydrodynamics of the multistage fluidized bed reactor with and without solids are studied. The hydrodynamics consists of pressure drop and hold-up of the reactor for the different operating conditions. The pressure drops are measured for different gas-flow rates, solid flow rates and weir heights and found to match excellently with the correlation. The pressure drop and solids holdup decreases with increase in mass velocity of gas. The pressure drop is found to increase in particle density. A maximum deviation of 25% is observed between experimental and predicted values. The maximum energy lost in form of pressure drop at minimum mass velocity of gas, maximum solid flow rate (lime particles) and maximum weir height is around 555 N/m<sup>2</sup>.
- 2. The solids holdup increases with respect to weir height and is found in the range of 0.067 to 0.159 kg for lime particles.
- 3. The mean holding time for the particles increases in increase in mass velocity of gas. This indicates suitability of system when large residence times are required.
- 4. At a particular solid flow rate, the height of the solids in the downcomers deceases when mass velocity of gas was increases. At a particular mass velocity of gas, the height of the solids in the downcomers increases when solids flow rate is increased. The height of the solids in the downcomers increases with time indicating flooding of the bed. Downcomer plays an important role in stable operation of the reactor.
- 5. The hydrated lime particles fluidizes with a tendency for agglomeration, which requires addition of some extraneous particles to improve fluidization quality.

# **CHAPTER 4**

# HYDRODYNAMICS OF MIXED PARTICLE SYSTEM IN A MULTISTAGE FLUIDIZED BED REACTOR

# Hydrodynamics of mixed particle system in a multistage fluidized bed reactor

#### 4.0 Introduction

Fluidized-bed based devices provide stable operation and intense mixing. Mixing of solids in a fluidized bed seems to be attractive since there are no moving parts in the separator, considerable reduction in space, simplicity in construction, operation and maintenance, consistency of products, easy installation, etc. The important characteristics of fluid-like flow behavior of solids in gas–solid fluidized beds have been widely recognized and is used to analyze fluidized-bed behavior as the flowability of gas–solid mixtures [Rowe and Nienow, 1972] and particle mixing and segregation [Hoffmann et al., 1993; Solimene et al., 2004; Solimene et al., 2006; Bokkers et al., 2004; Hartholt, 1997; Palppan and Sai, 2008].

It is well known that fluidization can be divided into two classes: aggregative fluidization and particulate fluidization [Wilhelm and Kwauk, 1948]. In general, aggregative fluidization appears in gas-solids systems. Its characteristic is that particle distribution in the gas is not uniform: there exist gas bubbles and solid agglomerates, because contact between gas and solids is not good, heat and mass transfer rates are low.

In order to increase reaction rate of gas-solid fluidization, many researchers have sought for methods for particulatization of aggregative fluidization. (the word "particulatization" gives the sense of converting aggregative behavior into particulate fluidization through "homogenizing" particle distribution in fluid. Although most of the literature dealing with fluidization of powders has been developed from the study of mono-disperse beds of particles, practical applications of fluidization technology are often concerned with mixtures of solids differing at least in density whenever a wide distribution of particles is present, if not also in size and shape, as in the case of processes which use more than one particulate species. Both in fixed and fluidized bed applications, however, important properties of a multicomponent bed can hardly be related to those of individual solids. Experimental results show that the process of fluidizing ultrafine particles usually involves slugging, channeling, disrupting and agglomerating. Experiments demonstrate further that when particles agglomerate during fluidization, there exists a fixed-bed region of large agglomerates at the bottom, a fluidized region of smaller agglomerates in the middle and a dilute-phase region of even smaller agglomerates, including discrete, unassociated particles, further up in the fluidized bed [Honhzhong et al., 2003].

In the Chapter-3, investigations on the hydrodynamics of MFBR with lime particles have been discussed in details. Since the main objective of this study is to remove sulfur dioxide from flue gases in the reactor under stable and uniform fluidized bed condition. It was concluded that lime being relatively soft and hygroscopic in nature, agglomeration of particles was observed, which played an important role in controlling particle motion through the downcomer resulting in choking and blockage of path for solids top flow. Agglomeration refers to cohesion of particles to each other due to Vander-Waal force of attraction, reducing the fluidization of particles to some extent.

The addition of extraneous particles into beds could weaken cohesive forces, reduce the natural agglomerate sizes, and improve fluidization quality [Wang et al., 1998]. Good fluidization may result from mixing two similar kinds of particles with different particle densities. There will be good fluidization quality if the additive particles tend to loosen the structure of agglomerates and lead to high porosity.

In the present investigation, to avoid typical agglomeration, a dolomite is introduced as the most preferred solid with lime as it is cheap and easily available without altering the reaction rate. Literature survey reveals that use of dolomite creates no problem in its fluidization and it is also easily available and not expensive [Kato et al., 1994]. But, when solids with different densities and sizes are mixed, a high degree of segregation may appear, with the dolomite settling at the bottom of the bed (jetsam) and the lime rising to the top (flotsam). For a given size of dolomite and lime, the achieving of a good degree of mixing depends on the relative concentration of both solids in the bed and also on the gas velocity, this being related to the minimum fluidization velocity for the mixture. The unique features and advantages of multistage fluidized bed reactor (MFBR) made it attractive both for catalytic and not catalytic reactions. The common features in each group of applications are that they are operated with bed solids that are dissimilar both in sizes and densities. Most of the studies reported in the literature were based on single particle systems and with narrow size group of particles. Therefore, in the present studies an attempt has been made to study the hydrodynamic characteristics of a MFBR with mixed type of particles (binary systems) of same sizes and different densities. Before conducting the

hydrodynamic study of binary mixture, the minimum fluidization velocity of the mixture has to be calculated to ensure the stable range of operation.

# 4.1 Minimum fluidization velocity of a binary mixture

The attempts to relate the minimum fluidization velocity of a binary mixture of particles of different sizes and/or densities to the properties of its individual solid components have generally been based on the extension of equations originally developed for predicting the  $u_{mf}$  of a mono-disperse bed. Ergun (1952) proposed the correlation of pressure drop through the particle bed as follows:

$$\frac{\Delta P_s}{H} = \frac{(1-\varepsilon)}{\phi_s d_p \varepsilon^3} \left[ \frac{150(1-\varepsilon)\mu_g u_g}{\phi_s d_p} + 1.75\rho' u_g^2 \right]$$
(4.1)

This correlation can be applied over the wide velocity range, that is, from laminar to turbulent regions. As the shape factor and voidage in this correlation can hardly be determined, Wen and Yu (1966b) modified Eq. (4.1) by eliminating these factors and obtained the equation for the minimum fluidization velocity for a single-component system as follows:

$$Ar = 24.5R_{e_{mf}}^2 + 1650R_{e_{mf}}$$
(4.2)

where,

$$Ar = \frac{d^{3}\rho'(\rho - \rho')g}{\mu^{2}} \text{ and } R_{e_{mf}} = \frac{d\rho' u_{mf}}{\mu}$$
(4.3)

In order to apply Eq. (4.2) to binary systems, it is necessary to define the particle diameter and the density of the binary system. For this purpose, it was assumed that the 'mixture density' is related as

$$\frac{1}{\rho} = \frac{w_F}{d_F} + \frac{w_P}{d_P}$$
(4.4)

and found that the smallest deviation between experimental and calculated values of  $u_{mf}$  was obtained by adopting a 'mixture diameter' definition such as to give the same total surface area per unit weight of the binary system:

$$\frac{1}{d_p} = \frac{w_F}{d_F \rho_F} + \frac{w_P}{d_P \rho_P}$$
(4.5)

Analogously, starting from the general form of Eq. (4.2), that is,

$$Ar = AR_{e_{mf}}^2 + BR_{e_{mf}}$$
(4.6)

where,

....

$$A = 36.2 \left(\frac{d_P}{d_F} \frac{\rho_F}{\rho_P}\right)^{-0.196}$$
(4.7)

As for parameter B, it is necessary to consider several exceptional cases and the following two equations are obtained.

$$B = 1397 \left(\frac{d_p}{d_F} \frac{\rho_F}{\rho_P}\right)^{0.296}$$
(4.8)

$$B = 6443 \left(\frac{d_P}{d_F} \frac{\rho_F}{\rho_P}\right)^{-1.86}$$
(4.9)

Palappan and Sai (2008) concluded from their experiment that the minimum fluidization velocity for the particles of different sizes and densities depends strongly on the mixing condition of the bed and on the volume fraction of the fluidizing media. The correlation to estimate the true minimum fluidization velocity at which both coarse and fine particles are completely fluidized for binary component systems is given by Eq. (4.6) with Eqs. (4.7) and (4.8) for completely mixed beds and Eqs. (4.7) and (4.9) for partially mixed beds.

#### 4.2 Experimental set-up and techniques

## 4.2.1 Materials

The feed was a binary mixture of solid particles of same size, but different density. All the particles selected for the study were in the Geldart group B class. Reasonably same sized particles were obtained for each mixture by taking a single screen cut using the 'BIS' sieves. Two sets of experiments were carried out with the solid particles in different proportion. In the first set, a mixture of dolomite (25%) and lime (75%) particles of size (426  $\mu$ m) were used and in the second set, mixture of dolomite (50%) and lime (50%) particles of same size(426  $\mu$ m) were used. The ratio of the density of the heavier to lighter particles was more than 1.35 for both the types of particle. Table 4.1 represents the properties of the solids used in the present study. In both the set variation of pressure drop with gas velocity were studied with different solid flow rate and different weir height.

Charactristics	Lime (lighter)	Dolomite (heavier)	
Average diameter (µm)	426	426	
Size range	+500-353	+500-353	
Sphericity	0.7	0.7	
Particle density(kg/m <sup>3</sup> )	2040	2700	
Geldart group	В	В	
<sup>a</sup> Terminal velocity(m/s)	3.79	5.43	
Minimum fluidization velocity (m/s)	0.112	0.120	

Table 4.1. Physical properties of solids used in the present study

<sup>a</sup>Value calculated according to Haider and Levenspiel (1989)

The minimum fluidization velocity of mixed particle system was calculated using the Eq. (4.6) with Eqs. (4.7) and (4.8) and the velocity was found to be 0.118 m/s and 0.143 m/s for mixture of dolomite (25%) and lime (75%) particles and dolomite (50%) and lime (50%) particles respectively. Since the minimum fluidization velocity of mixture was not much higher compared to minimum fluidization velocity of individual components, the pressure drop studies of mixed particle system was investigated under the same set of operating conditions pursued for single particle system. The objective of the study was to get rid of the agglomeration of lime particles using binary mixtures.

# 4.2.2 Description of the set up and procedure

The schematic of the experimental setup as shown in Fig. 2.6 which was used to study the hydrodynamics of single particle system has been used for mixed particle system and discussed in Chapter-2. Air at a particular flow rate was introduced into the empty column, after choosing a composition of the solid mixture. Premixed solids were introduced by solid feeder into the bed through the downcomer at specified solids flow rate by gravity flow. The same operational procedure was followed as done in case of single particle system. The system was allowed to attain the steady state. The steady state was verified by checking the inflow and outflow of the material into column. The pressure taps at each stage of the reactor and entire column were provided and connected to U-tube manometers by silicon tubes in order to measure the differential pressure at each stage and total pressure drop across the entire column. The pressure differences ( $\Delta Ps$ ) for each gas-solids rate at each stage and entire column were measured experimentally according to equation 3.1(Chapter-3). All the experiments were carried out varying gas flow rate, solid flow rate and weir height. The operating variables considered for multi-particle system are given in Table 4.2.

Table 4.2. The operating range of variables and the experimental conditions	

Parameters	Values
Ambient Temperature (K)	$308 \pm 3$
Mass velocity of gas (kg/m <sup>2</sup> ·s)	$31 \times 10^{-2} - 56 \times 10^{-2}$
Mass velocity of solids (kg/m <sup>2</sup> ·s)	35.4 x 10 <sup>-3</sup> – 141.5 x 10 <sup>-3</sup>
u <sub>op</sub> /u <sub>mf</sub>	2.5 - 4.5
Weir height (m)	0.04 - 0.06

# 4.3 Pressure drop studies for mixed particle system (mixture of 25% dolomite and 75% lime by weight)

The grid plate pressure drop in the absence of the solids has been measured at different gas flow rates and the variations of pressure drop with gas velocity have been shown in Fig. 2.11 (Chapter 2). While operating the system with multi-particle solids, it was observed that all the stages of the reactor were identical in their operation as well as performance and no occurrence of flotsam and jetsam were observed in the column which may be due to slight difference in density of binary mixtures. Flow of the solids was also visualized through the Perspex column and no segregation of particles flow structures was recorded. Further it was observed that all the stages of the reactor were identical in their operation as well as performance and the pressure drops across each stage and across the entire column. No discernible difference in the pressure drop across the stage was noticed from stage to stage. In view of identical performance, the pressure drop due to solids across each stage was obtained from the difference between the pressure drop with and without solids. The variations of pressure drop across a single stage under various flow conditions was investigated and discussed in the following section.

## 4.3.1 Results and discussion

### Effect of weir height on pressure drop

The variation of pressure drop with the different weir height for a mixture of dolomite and lime in 25%, 75% proportion is shown in the Figs. 4.1 to 4.3. Figs. 4.1 to 4.3 indicate the effect of superficial mass velocity of gas on pressure drop at different weir heights at particular solid flow rate where pressure drop decreased with increase in gas velocity. Similar observation has been reported by Mohanty et al. (2008a). This may be due to the fact that the gas experiences energy loss in the form of pressure drop while passing through bed. The increase in pressure drop may be due to more particles–gas interaction as particle concentration increases due to increase in bed volume. The minimum pressure drops occurred in the column at high solid flow rate (84.7 x  $10^{-3}$  kg/m<sup>2</sup> ·s) were 62.0, 71.1 and 85.2 N/m<sup>2</sup> at 0.04, 0.05 and 0.06 m weir height respectively. The maximum pressure drops occurred in the column at minimum solid flow rate (42.3 x  $10^{-3}$  kg/m<sup>2</sup> ·s) were 96.0, 110.0 and 130.3 N/m<sup>2</sup> at 0.04, 0.05 and 0.06 m weir height respectively. The total column pressure drop at minimum solids flow rate was found to be in the range of 168 to 390 Pa and at maximum solids flow rate 186 to 455 Pa.



Fig. 4. 1. Effect of superficial mass velocity of gas on pressure drop at different weir heights and  $G_S = 42.3 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 4. 2. Effect of superficial mass velocity of gas on pressure drop at different weir heights and  $G_s = 63.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 4. 3. Effect of superficial mass velocity of gas on pressure drop at different weir heights and  $G_s = 84.7 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 

# Effect of superficial mass velocity of gas on pressure drop

Figs. 4.4 to 4.6 describe the effect of superficial mass velocity of gas on pressure drop at a particular weir height corresponding to a different mass velocity of solids. It was observed that the pressure drop decreased due to loss of kinetic energy with increase in mass velocity of gas at a particular weir height. This may be due to the fact that the decrease in pressure drop is due to decrease in particle concentration in bed as increase in mass velocity of gas increases the movement of particles leading to its discharge through downcomer. Similar observation was reported by Mohanty et al. (2008a). The pressure drop was minimum at minimum weir height and maximum at maximum weir height. The minimum and maximum pressure drop at minimum pressure drop at minimum weir height was 56.0 and 114.0 N/m<sup>2</sup> respectively. The minimum and maximum at maximum weir height was 56.0 and 114.0 N/m<sup>2</sup> respectively. The minimum and maximum pressure drop at maximum weir height was 79 and 151.2 N/m<sup>2</sup> respectively. The total column pressure drop at minimum weir height from 237 to 453 Pa.



Fig. 4. 4. Effect of superficial mass velocity of gas on pressure drop at different superficial mass velocity of solids and  $h_W = 0.04$  m



Fig. 4. 5. Effect of superficial mass velocity of gas on pressure drop at different superficial mass velocity of solids and  $h_W = 0.05$  m



Fig. 4. 6. Effect of superficial mass velocity of gas on pressure drop at different superficial mass velocity of solids and  $h_W = 0.06$  m

# Effect of superficial mass velocity of solid on pressure drop

Figs. 4.7 to 4.9 indicate the effect of superficial mass velocity of solids on pressure drop at a particular weir height corresponding to different mass velocity of gas. It was observed that pressure drop increased with increase in mass velocity of solids. This may be due to the fact that at higher mass velocity of solids at a particular mass velocity of gas decreases the hydrodynamic resistances within gas-solid system at the expense of gas motive pressure in form of pressure loss. At a particular solid flow rate, the pressure drop was maximum at minimum gas flow rate and minimum at maximum gas flow rate. Mohanty et al. (2008a) has reported similar observations.



Fig. 4. 7. Effect of superficial mass velocity of solids on pressure drop at different gas velocities and  $h_W$  =0.04 m



Fig. 4. 8. Effect of superficial mass velocity of solids on pressure drop at different gas velocities and  $h_W$  =0.05 m



Fig. 4. 9. Effect of superficial mass velocity of solids on pressure drop at different gas velocities and  $h_W$  =0.06 m

# 4.4 Pressure drop studies for mixed particle system (mixture of 50% dolomite and 50% lime by weight)

#### 4.4.1 Results and discussion

The variation of pressure drop with the different operating variables for a mixture of dolomite and lime in 50% proportion is shown in the Figs. 4.10 to 4.18.

# Effect of superficial mass velocity of gas on pressure drop

Figs. 4.10 to 4.12 indicate the effect of superficial mass velocity of gas on pressure drop at different weir heights at particular solid flow rate. The pressure drop decreased with increase in mass velocity of gas. As observed, the pressure drop due to this mixture was more than pressure drop due to mixture of 25% dolomite and 75% lime. Since density of dolomite is more, more frictional pressure drop is experienced. Similar observation has been reported by Mohanty et al. (2008a). The minimum and maximum pressure drops occurred in the column at minimum solid flow rate (42.3 x  $10^{-3}$  kg/m<sup>2</sup> ·s) and at minimum weir height were 64.0 and 120.0 N/m<sup>2</sup> and at maximum solid flow rate (84.7 x  $10^{-3}$  kg/m<sup>2</sup> ·s) and at minimum meir height were 65.0 N/m<sup>2</sup>. The minimum and maximum pressure drops occurred in the column at minimum weir height were 83.0 and 155.0 N/m<sup>2</sup> and at maximum solid flow rate (84.7 x  $10^{-3}$  kg/m<sup>2</sup> ·s) and at maximum weir height were 117.0 and 173.0 N/m<sup>2</sup>.



Fig. 4. 10. Effect of superficial mass velocity of gas on pressure drop at different weir heights and  $G_s = 42.3 \times 10^{-3} \text{ kg/m}^2$ .s



Fig. 4. 11. Effect of superficial mass velocity of gas on pressure drop at different weir heights and  $G_s = 63.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 4. 12. Effect of superficial mass velocity of gas on pressure drop at different weir heights and  $G_s = 84.7 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 

# Effect of superficial mass velocity of solids on pressure drop

Figs. 4.13 to 4.15 represent the effect of superficial mass velocity of solids on pressure drop at different mass velocity of gas. The pressure drop decreased with increase in mass velocity of gas at constant mass velocity of solids due to decrease in solids concentration. The minimum and maximum pressure drops occurred in the column was 64 and 143  $N/m^2$  at 40 mm weir height and 71 and 151  $N/m^2$  at 50 mm weir height 83 and 178  $N/m^2$  at 60 mm weir height. The energy spent in form of total column pressure drop at minimum weir height was found to be in the range of 192 to 429 Pa and at maximum weir height 249 to 534 Pa.



Fig. 4. 13. Effect of superficial mass velocity of gas on pressure drop at different solid flow rates and and  $h_W = 0.04$  m







Fig. 4. 15. Effect of superficial mass velocity of gas on pressure drop at different solid flow rates and  $h_W = 0.06$  m

# Effect of superficial mass velocity of gas on pressure drop

Figs. 4.16 to 4.18 describe the effect of superficial mass velocity of gas on pressure drop at a particular weir height corresponding to a different mass velocity of solids. It was observed that with increasing gas velocity the pressure drop decreases. This is due to the fact that as increasing in mass velocity of gas decreases the gas holdup, thus decreasing in frictional resistance between gas-solid and hence decreases the pressure drop. The minimum and maximum pressure drop occurred at weir height of 0.04 m was 67.0 and 143.0 N/m<sup>2</sup> respectively. Similarly the minimum and maximum pressure drop at weir height of 0.05 m was 83.0 and 178.0 N/m<sup>2</sup> respectively.



Fig. 4. 16. Effect of superficial mass velocity of solids on pressure drop at different gas velocities and  $h_w = 0.04$  m



Fig. 4. 17. Effect of superficial mass velocity of solids on pressure drop at different gas velocities and  $h_W$  =0.05 m



Fig. 4. 18. Effect of superficial mass velocity of solids on pressure drop at different gas velocities and  $h_W$  =0.06 m

# 4.5 Development of correlations for pressure drop of mixed particle systems

An attempt has been made to correlate the friction factor with variables of the system. Conceivable variables on which the friction factor in the present system may depend are:

(a) flow properties- gas velocity (ug) and solids velocity (us);

(b) geometrical properties – height of the weir  $(h_w)$ , and diameter of mixed particles(dp);

(c) physical properties—namely the density of gas ( $\rho_g$ ), density of mixed solid ( $\rho_s$ ) and gravitational constant (g).

The friction factor thus becomes a function of following sensitive parameters, each of them trying to exert it influences:  $f = f[u_s, u_g, g, d'_p, h_w, \rho'_s, \rho_g]$ 

The variables grouped into dimensionless numbers by employing Buckingham's  $\pi$  theorem, which yields the following equation;

The dimensionless analysis is 
$$f = k \left[ \frac{u_s}{u_g} \right]^a \left[ \frac{gd_p}{u_g^2} \right]^b \left[ \frac{d_p}{h_w} \right]^c \left[ \frac{\rho_s - \rho_g}{\rho_s} \right]^d$$
 (4.10)

In order to establish the functional relationship between friction factor and the various dimensional groups in Eq. (4.10), multiple linear regression analysis [Douglas et al., 2001] has been performed to evaluate the constants and coefficients of the equation. The most closely related correlation on the statistical analysis which yields the minimum percentage error, presents the best possible correlation as follows:

$$f = 9.0 \left[ \frac{u_s}{u_g} \right]^{0.24} \left[ \frac{gd'_p}{u_g^2} \right]^{1.12} \left[ \frac{d'_p}{h_w} \right]^{0.2} \left[ \frac{\rho'_s - \rho_g}{\rho'_s} \right]$$
(4.11)

The correlation coefficient and the standard deviation of the experimental data from regression analysis are found to be 0.98 and 1.23 respectively. The predicted values of friction factor ( $f_c$ ) from Eq. (4.11) have been plotted against the experimental values ( $f_c$ ) in Fig. 4.19, where the solid line represents the regression equation and points are predicted values. The empirical correlation satisfies the experimental data of the present system satisfactorily.



Fig. 4.19. Comparison of the experimental and predicted friction factor in multistage counter-current fluidized bed reactor

## 4.6 Effect of aspect ratio on distributor to bed pressure drop ratio

Figs. 4.20 to 4.22 present the effect of aspect ratio on distributor to bed pressure drop ratio (R) at a particular solids velocity corresponding to different operating gas velocity for the first set of bed material (25% dolomite and 75% lime). Since major objective was to use higher percentage of lime particles to remove SO<sub>2</sub>, only first set of bed materials were analyzed to verify the improvement in the uniform fluidization. The same trend emerged as observed in case of single particles. But there was no sharp increase in R while decreasing the aspect ratio indicating improvement in quality of fluidization .The linearly increase of R suggests the decrease in agglomeration of particles and uniform distribution of particles in the bed.



Fig. 4. 20. Effect of aspect ratio on distributor to bed pressure drop ratio for mixed particle system and  $G_S = 42.3 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 4. 21. Effect of aspect ratio on distributor to bed pressure drop ratio for mixed particle system and  $G_S = 63.5 \times 10^{-3} \text{ kg/m}^2 \text{ s}$ 



Fig. 4. 22. Effect of aspect ratio on distributor to bed pressure drop ratio for mixed particle system and  $G_s = 84.7 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 

#### 4.7 Summary of the findings

The mixing behavior of a binary mixture differing in particle density with the same size have been studied for two set of feed mixtures and the results are reported through of a novel approach. The column pressure drop for two feed mixture are obtained. The summary of results was as follows;

- 1. The formation of fluidized beds in all stages of a multistage fluidized bed reactor is not immediate and the stages fill-up in sequences from top to bottom and the approach to steady state is arrived in sequence in the system. A minimum solid flow rate is required to form a fluidized bed in each stage in the system.
- 2. The dolomite particles affect the pressure drop substantially for every operating variable in the multistage fluidized bed reactor. The maximum pressure drop occurred with the first set of particles, where dolomite proportion is 25%, is  $151.6 \text{ N/m}^2$  at minimum solid flow rate and maximum

gas flow rate. The maximum pressure drop occurred with the second set of particles, where dolomite proportion is 50%, is 178.8  $N/m^2$  at the same operating conditions. The increase in pressure drop is due to the increase in dolomite ratio, where dolomite is denser than that of lime.

- 3. For higher dolomite ratio, the no choking is observed in which the particles were freely flowing in the bed.
- 4. The increase in the gas flow rate increases the solid flow rates to the underlying stages of the multistage fluidized bed reactor through the downcomer. Thus, the increase in the gas flow rate decreases the bed height, leading to the decrease in the pressure drop.
- 5. Addition of extraneous particles improves the fluidization quality of the hydrated lime particles.

# **CHAPTER 5**

# REMOVAL OF SULFUR DIOXIDE IN A MULTISTAGE FLUIDIZED BED REACTOR

# Removal of sulfur dioxide in a multi-stage fluidized bed reactor

### 5.0 Introduction

Emission of SO<sub>2</sub> and NOx from stationary sources, primarily from power stations, industrial heaters and cogeneration plants, causes a major environmental problem. Both SO<sub>2</sub> and NOx are mainly produced by power stations. About 99% of the sulfur dioxide in air comes primarily from industrial activities that burn fossil fuels containing sulfur and its compounds. In addition zinc and copper roasting and smelting plant also emits sulfur dioxide. Sulfur dioxide is also present in the exhaust of automobiles due to the internal combustion of fuel. The highest concentrations of sulfur dioxide in the air are found around sulfuric acid and fertilizer plant, petroleum refineries, ore roasting and smelting industries and power stations. Burning coal that has lower sulfur content can reduce sulfur dioxide emission. Using less polluting fuels, particularly gaseous fuels free from sulfur compounds may also reduce emissions of sulfur dioxide. However, when source correction techniques are limited the alternate approach is to flue gas desulphurization (FGD) equipments designed to remove SO<sub>2</sub> from the effluent gases released by various plants.

Large amount of currency are being spent to on numerous desulphurization projects worldwide since the 1950s [Marten, 1977]. The desulphurization efficiency and the sorbent utilization of the dry FGD processes so far tested haven't reached the level of wet and semi dry scrubbers. The major reason is that the residence time of the  $SO_x$  sorbent is very short causing very low desulphurization efficiency in all the existing processes. Accordingly, development of low-cost and high efficiency dry scrubbing technologies is of great demand to meet the stringent regulations as prescribed by pollution control boards and other regulatory agencies. Dry scrubbing systems are attractive in principle as compared to wet scrubbers in terms of cost because they do not require water and reheating energy.

Chiang et al. (2003) reported the application of single stage fluidized bed adsorber integrated with fabric filter for removal of acidic gases from flue gas in an incinerator. The removal efficiency of the adsorber for  $SO_2$  was 48.8-73.5% and it reached over 94% after passing through the filter. Tarelho et al. (2005) reported the

influence of operational parameters on SO<sub>2</sub> removal by limestone during fluidized bed coal combustion with removal efficiencies in the range of 25-85%. The increased in bed temperature beyond 825  $^{\circ}$ C decreased the removal efficiency. Pisani et al. (2003) reported on the application of a continuously operated binary fluidized bed single stage reactor using dolomitic lime stone (24µm) and inert solids of 500-590 µm diameter for removal of SO<sub>2</sub> .The maximum gas removal fraction of 76% was achieved at a temperature of 800  $^{\circ}$ C, a Ca/S ratio of 3 and a velocity of 0.8 m/s. Pisani et al. (2004) used a continuously operated binary fluidized bed single stage reactor using dolomitic lime stone (9.1 µm) and inert solids of 500-590 µm diameter for removal of SO<sub>2</sub> .The maximum efficiency of 97.7% was achieved at a temperature of 700  $^{\circ}$ C, a Ca/S ratio of 3 and a velocity of 0.8 m/s. However, it was reported that slightly change in operating condition reduced the efficiency appreciably.

Chu and Hwang (2005) investigated the application of a internally circulating fluidized bed reactor using calcium sorbent (385  $\mu$ m) and inert solids of 438  $\mu$ m diameter for removal of SO<sub>2</sub>. The maximum efficiency of 100% was achieved at a temperature of around 30 <sup>O</sup>C at higher relative humidity and at initial concentration of 500 ppm. Jiang et al. (1995) and other researchers have studied the removal of sulfur dioxide emission in circulating fluidized bed reactor at humid conditions. It was reported that the risk of solid deposition inside the reactor is very high and relatively high removal efficiency are difficult to achieve by the reactor.

Critical appraisal of the literature survey reveals that to achieve high removal efficiency of  $SO_2$  use of multi-stage reactor seems to be attractive to meet the stringent regulations. Therefore, in the present investigation a multistage counter-current fluidized bed reactor have been designed and fabricated as a desulphurization apparatus. Some of the special features of this type of reactor are to enhance the performance of the reactor and at the same time it reduces the chances of channeling, narrows the residence time distribution of solids and thus approaches plug flow. The aim of the study is to analyze the operation of a three-stage fluidized bed reactor for sorption of  $SO_2$  gas on lime particles for a wide range of operating conditions.

#### 5.1 Preparation of sorbent

The sorbent used here lime (CaO) was obtained in powder form from commercially available chemical raw materials. After hydration, the cake was sun-

dried for three days and dried in an air oven at  $100 \pm 5$  <sup>o</sup>C for two hours before it was crushed and sieved to the desired size range of 426 µm. Table 5.1 represents the physical characteristics of adsorbents used in the present study. The physical characteristics such as Brunauer–Emmett–Teller (BET) specific surface area was measured with a Micromeritics surface area analyzer that utilized the BET lowtemperature N<sub>2</sub> adsorption technique. The chemical properties of the calcium sorbent measured by atomic absorption spectrometer (AAS), thermogravimetric analyzer (TGA) is shown in Table 5.2.

Adsorbent	Characteristic	Value
Lime	Average particle diameter (μm)	426
	Density (kg/m <sup>3</sup> )	2040
	Minimum fluidization velocity at 35 <sup>O</sup> C (m/s)	0.112
	Specific area of unreacted sorbent (m <sup>2</sup> /g)	15
	Average pore diameter $(A^0)$ Pore volume (cc/gm)	93.4 1.28

Table 5.1. Physical characteristics of hydrated lime

Table	5.2.	Average chemical	composition	of h	ydrated l	ime
-------	------	------------------	-------------	------	-----------	-----

Compound	Weight (%)		
СаО	25.0		
Ca(OH) <sub>2</sub>	64.4		
CaCO <sub>3</sub>	4.76		
MgO	3.22		
Impurities	2.62		

# 5.2 Experimental apparatus and procedure

Fig. 5.1 is the schematic of the multi-stage fluidized bed reactor, which was used for hydrodynamic studies as discussed in Chapter 3, Fig. 3.1, has been used with provision of supplying the air-SO<sub>2</sub> mixture to the system. In order to generate synthetic air-SO<sub>2</sub> mixture, in composition similar to that of the exhaust of sulfuric acid plants, cupper smelters was made by mixing compressed air from an air compressor and SO<sub>2</sub> gas from an SO<sub>2</sub> gas cylinder.



Fig. 5. 1. Schematic diagram of the experimental set-up of a three-stage countercurrent fluidized bed reactor
Provision was made to feed the air-SO<sub>2</sub> mixture at the base of the fluidized bed reactor. The air-SO<sub>2</sub> mixture was generated by mixing air and SO<sub>2</sub> in an air-jet ejector assembly for uniform gas composition. Compressed air from the compressor was used as the motive fluid in the ejector to aspirate and thoroughly mix air with SO<sub>2</sub> from the SO<sub>2</sub> gas cylinder. Fig.5.1a is the schematic of the ejector used for gas mixing. The ejector was mounted with a downward slope of 30° with an air nozzle perfectly aligned along the axis of the ejector throat to ensure an axially symmetrical jet. The air nozzle was fixed at a projection ratio (which is the ratio of the distance between the nozzle tip and the beginning of the parallel throat to the throat diameter), by 3.78, which was determined experimentally for obtaining the highest possible mass ratio of aspirated gas. Compressed air at the desired pressure and flow rate was forced through the air nozzle and regulated by a valve. Simultaneously the SO<sub>2</sub> was routed at a controlled rate through SO<sub>2</sub> gas regulator and into the ejector. The air-SO<sub>2</sub> gas mixed intensely in the mixing throat of the ejector and the mixture was fed into chamber fitted at the bottom of the column. Pre-calibrated rotameter were used to measure the gas flow rate.

Experiments were conducted by setting the gas flow rate of  $31.27 \times 10^{-2}$  to  $56.4 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s}$  corresponding to solid flow rate of  $71.0 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$  and  $141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ . The weir height of the downcomer was kept at  $3 \times 10^{-2}$  m and  $7 \times 10^{-2}$  m and the gap between the downcomer bottom and the grid plate were kept as 0.015 m and 0.03 m respectively. For each gas flow rate, the inlets SO<sub>2</sub> loadings were varied from 500 to 1500 ppm in three stages. The study was carried out at room temperature and a pressure of 1 atmospheric absolute. Fig. 5.1b is the schematic of the gas sampling arrangements. Percentage removal efficiency of SO<sub>2</sub> was calculated for each experimental run by the formula.

$$\eta_{SO_2} = \frac{c_{in,so_2} - c_{out,so_2}}{c_{in,so_2}} \times 100$$
(5.1)

The removal efficiency of SO<sub>2</sub> in stage i, can be calculated as,

 $\eta_{SO2} = (C_{i+1} - C_i) / C_{i+1}$ (5.2)

Where  $C_i$  and  $C_{i+1}$  are outlet and inlet sulfur dioxide concentrations in gas.



Fig. 5.1 (a). Schematic diagram of ejector



Fig. 5.1 (b). Schematic diagram of SO<sub>2</sub> gas connection and sampling arrangements

#### 5.2.1 Sampling and analysis

Prior to sampling of the gas, the reactor was operated for sufficient some time till all the stages of the reactor were identical in their operation and the pressure drops across each stage was almost equal indicating steady and stable operation of reactor. Samples at the inlet of the column and outlet of each stage were drawn at flow rate of 1.0 LPM. Under steady state operating conditions, the SO<sub>2</sub> gas samples were collected at each point with the help of midget impinges and aspirator bottles. The gas samples were analyzed for sulfur dioxide by the "Tetrachloro-Mercurate method" [IS: 5182(Part-VI). The method consists of passing a portion of the air sampled, through a solution of absorbing medium (sodium tetracholoromercuarte) and analyzing the resulting solution spectrophotometrically by UV-visible recording spectrophotometer.

Absorbing solution of 0.1M sodium tetrachloromercurate was prepared by dissolving 27.2 gm (0.1 mole) mercuric chloride and 11.7gm (0.2 mole) sodium chloride in 1.0 liter of distilled water. This solution could be stored at room temperature for several months. The indicator, p-rosaline hydrochloride of 0.04%(w/v), acid bleached was prepared by dissolving 0.20 gm of p-rosaline hydrochloride in 100ml of distilled water and filtering the solution after 48 hours. This solution was stable for at least three months when stored in the dark and kept cool. The p-rosaline was used to develop a purple color with an absorbance maximum at 560 nm. The p-rosaline should have an assay of 95% percent or better and 20 ml of this solution was pipetted into a 100 ml volumetric flask to which 6 ml of concentrated hydrochloric acid was added. This solution should be pale yellow with a greenish tint. It was stored at room temperature in an amber bottle for a week to use. When SO<sub>2</sub> from the air stream was absorbed in a sodium tetrachloromercurate solution, it formed a stable di-chlorosulphitomercurate complex. The amount of SO<sub>2</sub> was then estimated by the color produced when p-rosaline hydrochloride when added to the absorbing solution. The color was estimated by using a spectrophotometer from

Eq. (5.3).

SO<sub>2</sub> in 
$$\mu g/m^3 = \frac{\mu g \text{ of } SO_2 / ml x \text{ vol of absorbing reagent x 1000}}{\text{vol of air sampled in liters}}$$
 (5.3)

a calibration curve. The calculation of  $SO_2$  concentration in the sample is given by in

$$(SO_2 \text{ in ppm} = SO_2 \text{ in } \mu g/m^3 \times 3.82 \times 10^4)$$

For the preparation of calibration curve, a dilute solution of sodium metabisulphite having strength of 1  $\mu$ g of SO<sub>2</sub> per ml is required. The stock solution of sodium metabisulphite is diluted in appropriate proportion to obtain solution of SO<sub>2</sub> containing 1  $\mu$ g of SO<sub>2</sub> per ml. This solution was used as the working standard solution. Eight standard volumetric flasks of 10 ml capacity were taken and 0.0, 1.0, 3.0, 5.0, 7.0, 9.0, 10.0 ml the working standard solution of SO<sub>2</sub> per ml is added in the volumetric flasks and the volume is made up to 10ml by adding absorbing reagent. For UV spectrophotometric analysis 1ml of sulfamic acid, 2 ml of pararosaniline hydrochloride solution and 2 ml Formaldehyde solution (0.2%) is added to each flask. It is kept for 30 minutes for color development and the absorbance is measured with UV-VIS spectrophotometer with a maximum wavelength at 560 nm. The calibration curve is drawn by plotting the absorbance versus concentration of SO<sub>2</sub> in  $\mu$ g/ml of the sample as shown in Fig. 5.2.

The measurement has been reported to the nearest 0.005 ppm at concentrations below 0.15 ppm and to the nearest 0.01 ppm for concentrations above 0.15 ppm. Ozone and nitrogen dioxide interfere if present in air samples at concentrations greater than SO<sub>2</sub>. Interference of nitrogen dioxide was eliminated by including 0.06% sulfamic acid in the absorbing reagent. This may, however, result in a different calibration curve of lower sensitivity and in greater losses of SO2 on shortage of the sample for more than 48 hours after o-toludine subsequent to sample salts, interfere by oxidizing Heavy metals, especially iron collection. dichlorosulfitomercurate during sample collection. This interference was eliminated by including ethelenediaminetetracaetic acid in the absorbing reagent. Sulfuric acid or sulfate do not interfere .If large amounts of solids materials are present, a filter may be used advantageously upstream; however a loss of SO<sub>2</sub> may occur. In the adsorption experiments, detailed studies have been carried out to determine the effect of gas and solids flow rates, inlet loading of sulfur dioxide on the percentage removal of sulfur dioxide using lime as the scrubbing medium. The range of concentration for  $\mathrm{SO}_2$ monitoring was 500-1500 ppm with accuracy  $\pm 10$  ppm.

#### 5.2.2 Results and discussion

The effect of various operating parameters on removal efficiency of sulfur dioxide in the reactor was investigated and the effect was discussed below;



Fig. 5. 2. Calibration curve of SO<sub>2</sub> concentration at  $\lambda_{max} = 560$  nm

### Effect of inlet SO<sub>2</sub> loading on the percentage removal efficiency of SO<sub>2</sub>

Figs. 5.3 to 5.4 represent the percentage removal efficiency of SO<sub>2</sub> ( $\eta_{SO2}$ ) at different inlet SO<sub>2</sub> loading at different conditions. It may be seen from Figs. 5.3 and 5.4 that increasing in inlet SO<sub>2</sub> loading decreased removal efficiency of SO<sub>2</sub> at a particular solid velocity and weir height. Similar observation has been reported by Mohanty et al. (2008c). The maximum removal efficiency of sulfur dioxide for inlet concentration 500 ppm was 65% and 62% at 70 mm weir height with mass velocity of solids 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s and 31.2 x 10<sup>-2</sup> kg/m<sup>2</sup> ·srespectively. The maximum removal efficiency of sulfur dioxide for inlet concentration 1500 ppm was 53% and 41% at 70 mm weir height at same other operating conditions. The reason of such decreasing trend may be due to an increase in sulfite concentration on the surface of lime particle and formation of suphite monolayer which results in decrease of the sorbent activity. While increasing in the mass velocity of gas, the percentage removal of sulfur dioxide also decreased since solids hold up in the bed decreased thereby decreasing the probability of diffusion of gas to lime particles.



Fig. 5. 3. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.07$  m and  $G_S = 141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 5. 4. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.07$  m and  $G_S = 71.0 \times 10^{-3} \text{ kg/m}^2 \text{ s}$ 

Figs. 5.5 to 5.6 represents the percentage removal efficiency of SO<sub>2</sub> ( $\eta_{SO2}$ ) at different inlet SO<sub>2</sub> loading at 30 mm weir height. It may be seen from the figures that increasing in inlet SO<sub>2</sub> loading decreased removal efficiency of SO<sub>2</sub> at a particular solid velocity and weir height. Similar observation has been reported by Mohanty et al.(2008c)The maximum removal efficiency of sulfur dioxide for inlet concentration 500 ppm is 62% and 49% at 30 mm weir height with mass velocity of solids 141.5 x  $10^{-3}$  kg/m<sup>2</sup> ·s and 71.0 x  $10^{-3}$  kg/m<sup>2</sup>s respectively. The maximum removal efficiency of sulfur dioxide for inlet concentration 1500 ppm is 55% and 41% at 30 mm weir height at same other operating conditions. The reason of such decreasing trend may be due to formation of the product layer on lime particles which results in decrease of the removal efficiency. While increasing in the mass velocity of gas, the percentage removal of sulfur dioxide also decreased since solids hold up in the bed decreased thereby decreasing the probability of gas-particle interactions.



Fig. 5. 5. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.03$  m and  $G_S = 141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 5. 6. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.03$  m and  $G_S = 71.0 \times 10^{-3} \text{ kg/m}^2 \text{ s}$ 

#### Effect of superficial mass velocity of solids on the percentage removal of $SO_2$

Figs. 5.7 and 5.8 describe effect of mass velocity of solids on the percentage removal efficiency of SO<sub>2</sub> ( $\eta_{so2}$ ) at different weir heights and particular gas flow rate (43.9 x 10<sup>-2</sup> kg/m<sup>2</sup> ·s). It may be seen that increasing in mass velocity of solids increases sulfur dioxide removal efficiency. At 0.07 m weir height, the maximum removal efficiencies are 60.5% and 41% for inlet concentration of 500 ppm and 1500 ppm respectively. The minimum removal efficiencies are 48% and 33.5% for inlet concentration of 500 ppm and 1500 ppm respectively. At 0.03 m weir height, the maximum removal efficiencies are 55% and 39% for inlet concentration of 500 ppm and 1500 ppm and 1500 ppm respectively. The minimum removal efficiencies are 40.5% and 21% for inlet concentration of 500 ppm and 1500 ppm respectively. The results indicate that as the mass velocity increases, the solid hold in each stage increases. These increases in hold-up of solids augment to adsorb more quantity of SO<sub>2</sub> on lime particle. Thus the increasing superficial mass velocity of gas increases the removal efficiency of SO<sub>2</sub>. A similar tendency of increasing removal efficiency SO<sub>2</sub> also reported by Mohanty et al. (2008c).



Fig. 5. 7. Effect of superficial mass velocity of solids on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.07$  m and Ga = 43.9 x 10<sup>-2</sup> kg/m<sup>2</sup> ·s



Fig. 5. 8. Effect of superficial mass velocity of solids on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.03$  m and Ga = 43.9 x 10<sup>-2</sup> kg/m<sup>2</sup> ·s

## Effect of superficial mass velocity of gas on the percentage removal efficiency of $SO_2$

Figs. 5.9 to 5.12 shows the effect of mass velocity of gas on the percentage removal efficiency of SO<sub>2</sub> ( $\eta_{so2}$ ) at different SO<sub>2</sub> loading, mass velocity of solids and weir height. Figs. 5.9 and 5.10 indicate that increasing in mass velocity of gas decreased removal efficiency at mass velocity of solids (141.5 x 10<sup>-3</sup> kg/m<sup>2</sup>·s and 71.0 x 10<sup>-3</sup> kg/m<sup>2</sup>·s) and weir height (0.07 m) due to decrease in concentration of solids at bed. Although, increasing in SO<sub>2</sub> concentration decreased the removal efficiency, but the difference of removal efficiency was more while increasing the concentration from 500 ppm to 1000 ppm than 1000 ppm to 1500 ppm. The reason of such decreasing tendency may be due to the sharp decrease of sorbent activity at lower range of concentration than higher range. Similar trends were also appeared for weir height 0.03 m as observed in Figs. 5.11 and 5.12. The removal efficiency was lowest at maximum mass velocity of gas at 56.4 x 10<sup>-2</sup> kg/m<sup>2</sup>·s, minimum mass velocity of solids at 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup>·s and minimum weir height of 0.03 m, which is 13.5% (Fig. 12). The reason of such lowest efficiency is due to decrease in residence time of gas and solids hold-up in the reactor system.



Fig. 5. 9. Effect of superficial mass velocity of gas on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.07$  m and Gs = 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s



Fig. 5. 10. Effect of superficial mass velocity of gas on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.07$  m and Gs = 71.0 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s



Fig. 5. 11. Effect of superficial mass velocity of gas on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.03$  m and Gs = 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> s



Fig. 5. 12. Effect of superficial mass velocity of gas on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.03$  m and Gs = 71.0 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s

#### Effect of weir height on the percentage removal efficiency of $SO_2$

Figs. 5.13 to 5.16 represents the effect of inlet SO<sub>2</sub> concentration on the percentage removal efficiency of SO<sub>2</sub> ( $\eta_{SO_2}$ ) at different weir heights. It can be seen from these figures that the percentage removal of SO<sub>2</sub> at the higher weir height was maximum as the solids reactant available was maximum. At 0.07 m weir height and mass velocity of solids of 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s , the highest and lowest removal efficiencies were 65% and 54% for 500 ppm inlet SO<sub>2</sub> concentration and 53% and 39% for 1500 ppm inlet concentration. At 0.07 m weir height and minimum mass velocity of solids of 71.0 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s, the range of removal efficiencies were 55% and 41% for 500 ppm inlet SO<sub>2</sub> concentration and 40% and 27% for 1500 ppm inlet concentration for the given range of mass velocity of gas . At 0.03 m weir height and mass velocity of solids of 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s, the removal efficiencies varied in the range of 62% and 47% for 500 ppm inlet SO<sub>2</sub> concentration and 47% to 31% for 1500 ppm inlet concentration for the given range of mass velocity of gas. At 0.03 m

weir height and mass velocity of solids of  $71.0 \times 10^{-3} \text{ kg/m}^2$  ·s, the removal efficiencies were 49% and 32% for 500 ppm inlet SO<sub>2</sub> concentration and 31% and 13.5% for 1500 ppm inlet concentration. As observed, increasing the weir height increased the removal efficiency due to increase in bed volume resulting in more gassolid interaction. However, the effect of weir height at lower concentration was not as much as observed at higher concentration indicating the presence of less quantity of reactive solids at lower height with increase in concentration. The difference of removal efficiency between two weir heights keeps on increasing with increase in SO<sub>2</sub> concentration.



Fig. 5. 13. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency at  $G_a = 31.2 \times 10^{-2} \text{ kg/m}^2$ .s and  $G_S = 141.5 \times 10^{-3} \text{ kg/m}^2$  ·s



Fig. 5. 14. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency at  $G_a = 56.4 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s}$  and  $G_s = 141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 5. 15. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency at  $G_a = 31.2 \times 10^{-2} \text{ kg/m}^2$  s and  $G_S = 71.0 \times 10^{-3} \text{ kg/m}^2$  s



Fig. 5. 16. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency at  $G_a = 56.4 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s}$  and  $G_s = 71.0 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 

#### Effect of temperature on the percentage removal of SO<sub>2</sub>

The experiment was carried out at the ambient temperature ranging from 35 to 42  $^{\circ}$ C. No discernible difference in the sulfur dioxide removal efficiency was observed due to change in temperature. This was similar to that observed by Ho and Shih (1992) in a fixed bed.

#### 5.3 Stage wise percentage removal efficiency of SO2

Figs. 5.17 to 5.20 indicate the percentage removal efficiency of  $SO_2$  at different stages at a particular solid flow rate, gas flow rate and weir height. It may be seen that as the solid reactant was fresh at the first stage, the maximum percentage of the inlet sulfur dioxide gets adsorbed and reacted at the surface of the calcium based sorbent (lime) for which removal efficiency was higher than other two stages. At the second stage, a low porosity product layer (CaSO<sub>3</sub>.0.5H<sub>2</sub>O) formed gradually on the surface of the sorbent and resulted in decrease in efficiency, which was similar to that observed by Ho and Shih (1992) in a fixed bed reactor. This may be due to the fact that since a low porosity product is formed, the diffusion resistance of SO<sub>2</sub> from the

emulsion phase to the inner untreated calcium sorbent gradually increases. Thus, the removal efficiency decreases. At the third stage, the removal efficiency of  $SO_2$  was quite low.



Fig. 5. 17. Effect of inlet SO<sub>2</sub> concentration on stage wise percentage removal efficiency at  $h_w = 0.07$  m and Gs = 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s



Fig. 5. 18. Effect of inlet SO<sub>2</sub> concentration on stage wise percentage removal efficiency at  $h_w = 0.07$  m and Gs = 71.0 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s



Fig. 5. 19. Effect of inlet SO<sub>2</sub> concentration on stage wise percentage removal efficiency at  $h_w = 0.03$  m and Gs = 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s



Fig. 5. 20. Effect of inlet SO<sub>2</sub> concentration on stage wise percentage removal efficiency at  $h_w = 0.03$  m and Gs = 71.0 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s

## 5.4 Theoretical determination of overall efficiency

The theoretical overall efficiency can be calculated in a multi-stage system, when the each stage efficiency is known by following way:

$$\eta_{T} = \eta_{1} + (1 - \eta_{1}) \eta_{2} + \{1 - \eta_{1} - (1 - \eta_{1}) \eta_{2}\} \eta_{3} + \dots + [1 - \eta_{1} - \dots - \eta_{n-1}] \eta_{n}$$
(5.4)  
where 'n' is the number of stages in a multi-stage fluidized bed reactor.

Since in the present investigation, three stages were used the overall removal efficiency may be expressed as :

$$\eta_{\rm T} = \eta_1 + (1 - \eta_1) \eta_2 + \{1 - \eta_1 - (1 - \eta_1) \eta_2\} \eta_3 \tag{5.5}$$

Fig. 5.21 represents a comparison between experimental and theoretical removal efficiency of SO<sub>2</sub>. From this figure it can be seen that theoretical value agrees well with experimental value with minimum percentage error. The correlation coefficient and the standard deviation of the experimental data from regression analysis are found to be 0.98 and 1.04 respectively. The deviation of the theoretical value from the experimental values is found to be within  $\pm 5$  % and it is presented in Fig. 5.22.



Fig. 5. 21. Comparison of experimental and theoretical percentage SO<sub>2</sub> removal efficiency



Fig. 5. 22. Deviation between experimental and theoretical percentage SO<sub>2</sub> removal efficiency

#### 5.5 Structural studies of samples by scanning electron microscope

It is well recognized that optical and electron microscopy is probably the best techniques for studying the structure of porous solid, though lengthy procedures are involved in sample preparation. The sulphation reactions with porous reactive solids often lead to pore mouth closing by product deposition and thereby the transport of reactant to the reactive interfacial surface is hindered. In order to examine such possibility, scanning electron microscope has been carried out for the materials before and after sulphation. The samples were coated with graphite suspensions and also with vacuum-evaporated gold-palladium alloy and examined under a scanning electron microscope operated at a potential of 15 kV. The microscope was used in the secondary electron mode. Figs. 5.23 and 5.24 are the SEM micrograph of hydrated lime before and after sulfation.

Fig. 5.23 shows the SEM micrograph of hydrated lime which was irregular in shape and porous. Fig. 5.24 shows the SEM micrograph of a particle of the sorbent reacted with  $SO_2$  at 1500 ppm. Examination of the reacted particles by SEM show that the desulphurization reaction changed their shape and surface. The surface of the particles reacted were covered by small grains. However the gas holes were still observed which were not sealed in the particles.



Fig. 5. 23. SEM micrograph of unreacted Ca(OH)<sub>2</sub>



Fig. 5. 24. SEM micrograph of Ca(OH)<sub>2</sub> reacted with SO<sub>2</sub> at 1500 ppm

#### 5.6 Summary of findings

The removal efficiency of  $SO_2$  in the multistage fluidized bed reactor is found to be a function of inlet concentration of  $SO_2$ , mass velocity of gas and solid and weir height. The experimental investigation shows that a comparatively high percentage removal efficiency of  $SO_2$  above 65 % can be achieved from a lean air– $SO_2$  mixture for 500 ppm inlet concentration at ambient temperature. The decrease in the efficiency with increase in  $SO_2$  concentration is mainly due to the increase in the sulfite concentration in the sorbent resulting in increase in diffusion resistance of  $SO_2$ from the emulsion phase to the inner untreated calcium sorbent. The results also indicate that, increase in solids flow rate increases the  $SO_2$  removal efficiency where as increase in the gas flow rate decreases the removal efficiency. This is due to the fact that increasing in gas flow rate decreases solids hold up and increasing in solids flow rate increases solids hold up. Further the increase in gas flow rate leads to reduction in the residence time of the  $SO_2$  gas molecules and hence there is a decrease in the efficiencies.

It was observed that the weir height has marginal influence on removal efficiency at lower concentration. The removal efficiency is higher at first stage than other stages due to availability of fresh solids. The maximum first stage removal efficiency was found to be 40% at mass velocity of gas of  $31.0 \times 10^{-2} \text{ kg/m}^2$  ·s and mass velocity of solids of 141.0 x  $10^{-3} \text{ kg/m}^2$  ·s and 70 mm weir height. This system shows that desired removal efficiency of SO<sub>2</sub> can be achieved to meet the emission standard prescribed by regulating agency irrespective of any temperature of flue gas.

An increase in  $SO_2$  removal efficiency is found to result from the increase of mean residence time of gas in the reactor, which indicates that either a decrease in superficial gas velocity or an increase in mass velocity of solids can make  $SO_2$  removal efficiency increase. For a given superficial gas velocity, it can prolong the residence time of flue gas in the reactor, which increases the contact time or reaction time between  $SO_2$  gas and the  $SO_2$  sorbents in the reactor. However, operating the system at a low gas velocity means that larger handling equipment must be used to achieve the same throughput. In addition, it should be noted that for any specific bed there is a minimum stable operating velocity and a maximum stable operating velocity, beyond which the reactor could not be normally operated steadily.

# **CHAPTER 6**

# THEORETICAL MODELING OF SULFUR DIOXIDE IN A MULTISTAGE FLUIDIZED BED REACTOR

### Theoretical modeling of sulfur dioxide in a multistage fluidized bed reactor

#### **6.0 Introduction**

Gas-solid fluidized bed reactors have found a wide range of industrial applications. However, the heat, mass transfer and reaction within such reactors are far from being well understood and effectively quantified due to complexity of gassolid interactions. Over the past three decades, several models have been developed with regard to the gas-solid fluidized bed. The purpose of each model is to analyze the gas-solid contact, delineate the exchange processes between the various phases as identified by each model, and study the effects of gas-solid properties and the operating velocity. The objective of a fluidized bed model is to combine the chemistry of the gas-solid reaction and several other hydrodynamic parameters mathematically to arrive at equations that are useful in estimating the degree of conversion and the size of the reactor.

Models pertaining to fluidized beds can generally be of two major types: models that consider only two phases and models that focus on a bubbling bed. In a two-phase model, one phase is constituted of bubbles, which are assumed to be devoid of solid particles, and the second is particle-rich dense phase or emulsion phase. In the simplest types (Level I) of two-phase models, the bubble-phase gas is attributed to the excess gas flow above that required to achieve minimum fluidization. In other words, the dense phase or the emulsion phase corresponds to a bed at the incipient state of fluidization. In a model of this type, parameters relating to bubble size variation or growth are not considered. When the bubble size parameters are considered to be either constant or adjustable, the model is designated Level II. In models designated Level III, bubble size variation with bed height and bed diameter is also considered.

The analysis of models and the inferences that can be drawn in terms of selection of a specific model for design applications is not a simple task. The main reasons are due to the many underlying principles that govern the development of models and the several assumptions that are not necessarily adopted in the same sense in all models. The models can be for a steady state or an unsteady state for either an isothermal or a non-isothermal reactor. The development of a model that covers the entire complex hydrodynamic, thermal, and chemical characteristics of the reactor is a challenging task for a fluidization engineer. It is also not a simple job to analyze the numerous models and select a specific one for universal application. The basics of fluidization and many fundamental parameters are under constant review by researchers, and refinements are continuously being suggested. Modeling concepts and selection criteria still constitute a frontier in fluidization research.

Bubbling fluidized beds have extensively been studied and variety of models of varying degree of complexity has been proposed in the literature [Davidson and Harrison, 1985; Johnson et al., 1986; Kuni and Levenspiel, 1991; McAuley et al., 1994; Kim and Choi, 1999; Hatzantonis et al., 2000; Lu et al., 2004; Harshe et al., 2004;; Alizadeh et al., 2004; Kiashemshaki et al., 2006]. Researchers have explained and made sense of the main features of the phases in fluidized beds and have been of great help in improving knowledge of interaction between the phases. Generally, for modeling of fluidized bed reactor either a two-phase model comprising of emulsion and bubble phases (Hatzantonis et al., 2000; Lu et al., 2004; Kiashemshaki et al., 2006; Harshe et al., 2004; McAuley et al., 1994) or a three phase model comprising of an additional cloud phase (Kim and Choi, 1999) is considered. For Geldart B type particles, at higher values of  $u_g/u_{mf}$  ratio, the presence of cloud phase can be considered negligible (Faltsi-Saravelou and Vasalos, 1991).

The mixing in a fluidized bed reactor is very complex in nature and difficult to characterize. In the literature both the phases (the emulsion and the bubbles) have been modeled either as a plug flow or perfectly mixed flow system. The assumption of plug flow for the bubble phase is usually valid; however, it is not clear at all whether the emulsion phase should be modeled as being perfectly mixed or in plug flow. Hymore and Laguerie (1984) successfully applied the two-phase model for the drying of moist air by alumina particles in a counter-flow multistage bubbling fluidized bed reactor. The model predicted in quite close with experimental results.

Neuzil et al. (1988) developed a model on two-stage counter-current periodically operated fluidized bed reactor for removal of  $SO_2$  from gases by active

soda. The model has not considered the hydrodynamic aspects of the reactor. Khani et al. (2008) simulated a bubbling fluidized bed reactor for the fluorination of uranium tetra-fluoride by fluorine gas by employing two phase models, with bubble phase assumed to be in plug flow, and the emulsion phase in plug flow and in perfectly mixed. The model calculations were compared with actual data in terms of fluorine conversion. The comparison of both model showed that the perfectly mixed model predicted the data satisfactorily.

The literature on modeling of a multistage bubbling fluidized bed reactor is very scanty. Therefore, it is necessary to develop a theoretical model for the prediction of hydrodynamic behavior of gas in a MFBR based on the assumptions as reported by Davidson and Harrison (1963) with some modification. Since literature suggests that the multistage fluidized bed reactor behaves as a plug flow reactor, in the present study both flow regimes (plug and perfectly mixed) were considered for the emulsion phase together with plug flow regime for the bubble phase, and were compared with experimental data.

#### 6.1 Model formulation in a multi-stage fluidized bed reactor

#### 6.1.1 Assumptions of the model

- I. The total bed consists of two phases i) Solid-free bubble phase ( $\epsilon = 1$ ) ii) Solid-rich emulsion phase ( $\epsilon = \epsilon_{mf}$ );
- II. All gas in excess of that required for minimum fluidization passes through the bed as bubbles;
- III. The bubble phase does not contain any solids;
- IV. The bubble phase is in plug flow and gas in bubbles is perfectly mixed;
- V. The bubbles are spherical, of constant size and evenly distributed ib the bed at any time;
- VI. As bubbles rise, bubbles exchange gas with rest of bed (emulsion phase. The inter-phase mass transfer results from two independent mechanism; bulk flow of gas and diffusion;
- VII. The mass transfer resistances between the particles and dense phase gas are neglected;

VIII. The solid particles are perfectly mixed in emulsion phase (The emulsion phase is perfectly mixed), but gas in emulsion phase is considered i) emulsion gas perfectly mixed (EGPM) model ii) emulsion gas in plug flow (EGPF) model;

Additional assumptions;

- I. Constant mean particle size is assumed through out bed;
- II. Emulsion of solids at top of bed is neglected;
- III. All the stages operate under same conditions of fluidization;
- IV. Holdup of solids is same on each stage;
- V. All the particles entering first stage present the same concentration in active species;
- VI. Constant mean particle size is assumed throughout the bed.

So the assumptions lead in fact to two distinct models; i) EGPM model ii) EGPF model. Fig. 6.1 and 6.2 are the schematic of both the model.



Fig. 6. 1. Schematic of EGPF Model



Fig. 6. 2. Schematic of EGPM Model

#### 6.1.2 Choice of a kinetic model

Borgwardt (1970) investigated on kinetics of the reaction of  $SO_2$  with calcined limestone and observed that at low temperature, the primary reaction in the flue gas desulphurization process is as follows;

$$Ca(OH)_2 + SO_2 \longrightarrow CaSO_3.0.5H_2O + 0.5 H_2O$$
 (6.1)

 $\Delta H^{0}_{298} = -238 \text{ kJ/mol}$ 

The reaction is first-order with respect to sulfur dioxide and calcium hydroxide. Rate constant has been expressed according to Arrehenius' law

$$k_1 = A_0 \exp\left(-\frac{E}{RT}\right) \tag{6.2}$$

where the activation energy of reaction is 32 kJ/mol, and for the pre-exponential factor we have  $A_0 = 2.314 \text{ m}^3/\text{mol.s}$  (Ghosh, 1987).

#### 6.1.3 Equations of the model

#### (a) Mass balance at the exit:

The mass balance for the reactant gas at the exit is

$$uC_{i} = u_{mf}C_{pi} + (u - u_{mf})C_{bi}(H)$$
(6.3)

Rearranging the terms, the concentration of gas leaving the ith stage,

$$C_{i} = \beta C_{bi}(H) + (1 - \beta) C_{pi}$$
(6.4)

Where  $\beta$  is the fraction of gas flow associated to the bubble phase, i.e.  $(\beta = 1 - \frac{u_{mf}}{v})$ .

#### (b) Mass balance of sulfur dioxide in bubble phase:

The mass balance on sulfur dioxide through the bubble phase of the i<sup>th</sup> stage is independent of the assumption relative to the behaviour of gas in emulsion phase. Consider an element of thickness dz, at a height of z in bed, containing N.dz bubbles. The material balance is written for a unit horizontal cross sectional area by considering total bed height H. Combining the inlet and outlet terms and rearranging, it gives

Rate of change of reactant concentration = Loss of reactant by exchange

$$N_{b}Q(C_{pi} - dC_{bi}) = N_{b}V_{b}u_{b}\frac{dC_{bi}}{dz}$$
(6.5)

Rearranging the Eq. (6.5), we get

$$\frac{dC_{bi}}{dz} \left( \frac{V_b u_b}{Q} \right) = (C_{pi} - C_{bi})$$
(6.6)

(c) Mass balance of sulfur dioxide in emulsion phase

#### *(i)EGPM Model*

The material balance is written for unit horizontal cross sectional area by considering total bed height H. Combining inlet and outlet terms;

$$\int_{0}^{H} N_{b}QC_{bi}dz + u_{mf}C_{i+1} = u_{mf}C_{pi} + C_{pi}\int_{0}^{H} N_{b}Qdz + \tilde{r}\int_{0}^{H} (1 - N_{b}V_{b})dz$$
(6.7)

Where the  $SO_2$  concentration in the emulsion phase, Cpi , is independent of height z .H is the height of expanded bed assumed equal to height of the down comer over the

distributor and  $\check{r}$  is the average rate of disappearance of SO<sub>2</sub> in the entire emulsion phase.

#### ii) EGPF Model

Let us now consider that the gas is in plug flow through the emulsion phase with all same assumptions. The material balance over a unit cross-sectional area of dense phase considering an infinitesimal height dz;

$$u_{mf}C_{pi} + N_bQC_{bi}dz = u_{mf}(C_{pi} + dC_{pi}) + N_bQC_{pi}dz + r_i(1 - N_bV_b)dz$$
(6.8)

Rearranging the term, we get

$$u_{nif} \frac{dC_{pi}}{dz} + N_b Q (C_{pi} - C_{bi}) + k_1 C_{pi} (1 - N_b V) dz = 0$$
(6.9)

Where the  $SO_2$  concentration in the emulsion phase, Cpi, and  $\check{r}$  are functions of z.

(d) Mass balance on the sorbent particles: At any time over the i<sup>th</sup> stage, the disappearance rate of calcium oxide (R) according to the rate kinetics can be written as:

$$\frac{\partial C_{Ri}}{\partial t} = -k_1 C_{Ri} C_{pi} \tag{6.10}$$

EGPM Model: The calcium hydroxide concentration is

$$C_{Ri} = C_{Rio} \exp\left(-k_1 C_{pi} \overline{\tau_i}\right) \tag{6.11}$$

Where  $\overline{\tau_i}$  is mean residence time of solids in each stage.

Which can be expressed as the function of initial concentration of sorbent,  $C_{R0}$  of the particles entering the first stage of the reactor:

$$C_{R} = C_{R0} \exp\left(-k_1 \sum C_{jj} \overline{\tau_j}\right) \qquad \text{for } j = 1 \text{ to } i \qquad (6.12)$$

EGPF Model: The sulfur dioxide concentration of gas varies throughout the emulsion phase, but the solid particles which are perfectly mixed, have at any time, an equal probability of contacting an element of gas volume, whose concentration  $C_{Pi}$  lies between  $C_{i+1}$  and  $C_{pi}(H)$ . Therefore, it can be accepted that considering all the particles, the sulfur dioxide concentration of gas is  $\overline{C_{Pi}}$  which is defined by the equation

$$\overline{C_{P_i}} = \frac{1}{H} \int_0^H C_{P_i} dz$$
(6.13)

(e) Sulfur dioxide concentration in the exit gas stream from  $i^{th}$  stage The rate of SO<sub>2</sub> disappearance in  $i^{th}$  stage can be expressed as

$$r_i = (k_1 \overline{C_{Ri}}) C_{Pi} \tag{6.14}$$

The term in the bracket remains constant over the entire emulsion phase so that the reaction rate can be considered as first order i.e.  $r_i = (k_1 \overline{C_R})C_{P_i} = kC_{P_i}$  (6.15)

#### (f) Analytical solution of model equation

#### (i) Bubble Phase

Integrating the Eq. (6.6) from 0 to H with boundary condition at z=0,  $C_{bi} = C_{i+1}$  and z=H,  $C_{bi} = C_{bi}(H)$ 

$$\int_{0}^{H} \frac{dC_{bi}}{(C_{pi} - C_{bi})} = \int_{0}^{H} \left(\frac{Q}{V_{b} u_{b}}\right) dz$$
(6.16)

$$C_{bi}(z) = C_{pi} + (C_{i+1} - C_{pi})e^{\frac{Qz}{u_b V_b}}$$
(6.17)

$$C_{bi}(H) = C_{pi} + (C_{i+1} - C_{pi})e^{-X}$$
(6.18)

Where  $X = \frac{QH}{u_b V_b}$  indicates the number of times a bubble is purged as it rises through the bed or number of times the gas within the bubble is exchanged with the particulate phase during the passage through the bed.

#### (ii) EGPM Model

Rearranging the terms of the Eq. (6.7) and simplifying, the equation is ;

$$u_{mf}(C_{i+1} - C_{pi}) = N_b Q \int_0^H (C_{pi} - C_{bi}) dz + \tilde{r} \int_0^H (1 - NV_b) dz$$
(6.19)

Eliminating  $C_{pi}$  from RHS of the Eq. (6.19), the equation is

$$u_{mf}(C_{i+1} - C_{pi}) = N_b u_b V_b \int_0^H dC_{bi} + \tilde{r} \int_0^H (1 - NV_b) dz$$
(6.20)

Integrating the Eq. (6.20) from 0 to H with boundary condition at z=0,  $C_{bi}=C_{i+1}$  and z=H,  $C_{bi}=C_{bi}(H)$ 

$$u_{mf}(C_{i+1} - C_{pi}) = N_b u_b V_b \{C_{bi}(H) - C_{i+1}\} + k C_{pi}(1 - N V_b) H$$
(6.21)

Putting the value of  $C_{bi}(H)$  in the Eq. (6.21) and rearranging, we get

$$C_{pi} = \frac{C_{i+1}(1 - \beta e^{-X})}{(1 - \beta e^{-X} + \frac{KH_{mf}}{u})}$$
(6.22)

$$C_{bi}(H) = C_{\mu} + (C_{i+1} - C_{\mu})e^{-X}$$
(6.23)

Putting the above values of  $C_{Pi}$  and  $C_{bi}(H)$  in the Eq. (6.4), it gives the SO<sub>2</sub> exit concentration from i<sup>th</sup> stage for the model.

#### (iii) EGPF Model

Eliminating  $C_{pi}$  from the Eq. (6.9) and simplifying it,

$$\frac{d^{2}C_{bi}}{dz^{2}} + \frac{u_{nf}\left\{1 + \frac{d}{dz}\left(\frac{u_{b}V_{b}}{Q}\right)\right\} + N_{b}u_{b}V_{b} + K_{1}(1 - N_{b}V_{b})\left(\frac{u_{b}V_{b}}{Q}\right)}{dz} \frac{dC_{bi}}{dz} + \frac{K_{1}(1 - N_{b}V_{b})}{Q}C_{bi} = 0 \quad (6.24)$$
Let us assume a = 
$$\frac{u_{nf}\left\{1 + \frac{d}{dz}\left(\frac{u_{b}V_{b}}{Q}\right)\right\} + N_{b}u_{b}V_{b} + K_{1}(1 - N_{b}V_{b})\left(\frac{u_{b}V_{b}}{Q}\right)}{\frac{u_{b}V_{b}u_{nf}}{Q}} \quad (6.25)$$

And 
$$b = \frac{K_1(1 - N_b V_b)}{\frac{u_b V_b u_{mf}}{Q}}$$
(6.26)

Now the Eq. (6.24) is

$$\frac{d^2C_{bi}}{dz^2} + a\frac{dC_{bi}}{dz} + bC_{bi} = 0$$
 is a second order differential equation with respect to C<sub>bi</sub>. Now

the equation can be written as

$$(D^2 + a D + b) C_{bi} = 0$$
 (6.27)

The auxiliary equation is  $m^2 + am + b=0$  (6.28)

Since this is a quadratic equation, the characteristic roots of the equation are  $m_1$  and  $m_2$ , then complementary functions of the equation=  $C_{bt} = c_1 e^{m_2} + c_2 e^{m_2}$  (6.29)

The above equation can be solved by following boundary equations;

At z=0 
$$C_{bi} = C_{i+1}$$
 and  $\frac{dC_{bi}}{dz} = 0$ 

At z = H,  $C_{bi} = C_{bi}(H)$ 

Differentiating the Eq. (6.29), we get

$$\frac{dC_{hi}}{dz} = c_1 m_1 e^{m_2} + c_2 m_2 e^{m_2 z}$$
(6.30)

At z =0, the equation becomes,  $C_{bi} = C_{i+1} = c_1 + c_2$  and  $c_1 m_1 + c_2 m_2 = 0$  (6.31)

At z =H, the equation (0 is 
$$C_{tx}(H) = c_1 e^{m_1 H} + c_2 e^{m_2 H}$$
 (6.32)

Solving the above equations, we get

$$c_1 = \frac{-m_2 C_{i+1}}{m_1 - m_2} \tag{6.33}$$

$$c_2 = \frac{m_1 C_{i+1}}{m_1 - m_2} \tag{6.34}$$

Putting the value of  $c_1$ ,  $c_2$  in the Eq. (6.32), the equation becomes

$$C_{bi}(H) = \frac{C_{i+1}}{m_1 - m_2} (m_2 e^{m_2 H} - m_2 e^{m_1 H})$$
(6.35)

Putting the value of  $C_{bi}$  in the Eq. (6.18), we get

$$C_{pi} = \left(\frac{C_{i+1}}{m_{1} - m_{2}}\right) \left[ m_{1i} \left(1 + \frac{H}{X} m_{2}\right) \exp(m_{2}H) - m_{2} \left(1 + \frac{H}{X} m_{1}\right) \exp(m_{1}H) \right]$$
(6.36)

Putting the values and solving a and b, these become,

$$a = \frac{X + K}{(1 - \beta)H} \tag{6.37}$$

$$b = \frac{K.X}{(1-\beta)H^2} \tag{6.38}$$

Where Ki is the number of reaction unit in emulsion phase

$$K_i = KC_{Ri} \frac{H_{mf}}{U}$$
(6.39)

Putting the values of a and b in the auxiliary Eq. (6.28), the equation becomes

$$H(1-\beta)m^{2} - (X+K)m + \frac{X.K}{H} = 0$$
(6.40)

Then the roots of the equation are

$$m_{1or2} = \frac{1}{2} \frac{X+K}{H(1-\beta)} \pm \frac{1}{2} \left[ \left( \frac{X+K}{H(1-\beta)} \right)^2 - 4 \frac{X.K}{H^2(1-\beta)} \right]$$
(6.41)

For i<sup>th</sup> stage

$$m_{\text{hor2}i} = \frac{1}{2} \frac{X + K_i}{H(1 - \beta)} \pm \frac{1}{2} \left[ \left( \frac{X + K_i}{H(1 - \beta)} \right)^2 - 4 \frac{X \cdot K_i}{H^2(1 - \beta)} \right]$$
(6.42)

$$C_{bi}(H) = \frac{C_{i+1}}{m_{1i} - m_{2i}} (m_{1i}e^{m_{2i}H} - m_{2i}e^{m_{1i}H})$$
(6.43)

$$C_{pi} = \left(\frac{C_{i+1}}{m_{i} - m_{2i}}\right) \left[m_{i}\left(1 + \frac{H}{X}m_{2i}\right) \exp(m_{2i}H) - m_{2i}\left(1 + \frac{H}{X}m_{i}\right) \exp(m_{i}H)\right]$$
(6.44)

Mean concentration of emulsion gas is given by

$$C_{pi} = \frac{1}{H} \left( \frac{C_{i+1}}{m_{ij} - m_{2i}} \right) \left[ \frac{m_{ij}}{m_{2i}} \left( 1 + \frac{H}{X} m_{2i} \right) \left[ \exp(m_{2i} H) - 1 \right] - \frac{m_{2i}}{m_{ij}} \left( 1 + \frac{H}{X} m_{ij} \right) \left[ \exp(m_{ij} H) - 1 \right] \right]$$
(6.45)

Putting the above values of  $C_{Pi}$  and  $C_{bi}(H)$  in the eqn.6.4, it gives the SO<sub>2</sub> exit concentration from i<sup>th</sup> stage for the model.

#### 6.1.4 Evaluation of parameters of model

(A) Parameters relating to bubbles:

(1) Number of bubbles: All the gas in excess of that required to incipiently fluidized the bed passes through the bed as bubbles, which are uniform in size and equally distributed throughout the bed. Then from the continuity of equation,

$$N_{b}V_{b}u_{b} = u - u_{mf} \tag{6.46}$$

Therefore Number of bubbles per unit bed volume=  $N_b = \frac{u - u_{mf}}{V_b u_b}$  (6.47)

(2) Bubble volume: 
$$V_b = \frac{\pi \overline{D}_b^3}{6}$$
 (6.48)

(3)Bubble diameter= $D_b(z) = D_{b0} + 2.048(u - u_{nf})^{0.94} z$  [Mori and Wen, 1975] (6.49)

(4)Initial bubble diameter= 
$$D_{b0} = 0.347 \left(\frac{A(u-u_{nf})}{n_d}\right)^{0.4}$$
 (6.50)

(5) Average bubble diameter = 
$$\overline{D_b} = D_{b0} + 2.048(u - u_{mf})^{0.94} \cdot \frac{H}{2}$$
 (6.51)

(6) Bubble rise velocity: The absolute rise velocity of a bubble  $(u_b)$  is the sum of natural rise velocity as given by Kunii and Levenspiel (1991) and the upward velocity of particulate phase between the bubbles.

$$u_{b} = (u - u_{nf}) + 0.711 \sqrt{(gD_{b})}$$
(6.52)

(B) Increase in bed height: The increase in bed height over that minimum fluidization is due to the presence of bubbles alone.

$$N_b V_b H = H - H_{mf} \tag{6.53}$$

Therefore 
$$H_{nf} = H(1 - N_b V_b) = H(1 - \frac{u - u_{nf}}{u_b})$$
 (6.54)

(C) Inter-phase mass transfer:

Every bubble, while rises through the bed, exchanges gas with emulsion phase. For a spherical void of diameter,  $D_b$ , the rate of exchange is attributed to a combination of "through flow" of gas, q given by Davidson and Harrison analysis (1963),

$$q = \frac{3}{4} u_{mf} \left( \pi D_b^2 \right) \tag{6.55}$$

And convective diffusion,

$$K_G = 0.975 D_G^{0.5} \left(\frac{g}{D_b}\right)^{0.25}$$
(6.56)

Where  $K_G$  is the mass transfer coefficient per unit surface area of bubble.

The total flow rate of gas between bubble and emulsion phase is, therefore

$$Q = q + K_G A = \left[\frac{3}{4}u_{mf} + 0.975 D_G^{0.5} \left(\frac{g}{D_b}\right)^{0.25}\right] (\pi D_b^2)$$
(6.57)

(D)Diffusivity of gaseous reactant in air= $D_g(T)=8.0X10^{-26}T^{4.5}$ [Treybal, 1980] (6.58)

#### 6.2 Results and discussion

The concentration of the gas stream exiting each stage has been calculated by solving the equation of the models. The calcium hydroxide concentration ( $C_{R0}$ ) of the sorbent particles entering the reactor, expressed in moles of calcium hydroxide per unit volume of solids, was determined by analysis. The developed mathematical model equations are simulated by MATLAB version 6.5 programme for the theoretical efficiency. The program was simulated for different variables and compared with experimental results.

## Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub>

Figs. 6.3 to 6.4 represent the effect of different inlet sulfur dioxide loading on the theoretical percentage removal of SO2 at a particular mass velocity of solids (141.5 x  $10^{-2}$  kg/m<sup>2</sup> ·s), particular mass velocity of gas (31.2 x  $10^{-2}$  kg/m<sup>2</sup> ·s). For this set of prediction, temperature was set to 310 K. It may be observed that the removal efficiency decreases as the inlet sulfur dioxide concentration increases. The percentage removal of SO<sub>2</sub> is higher for EGPF model compared to EGPM model. The percentage removal of sulfur dioxide is 62% for EGPF model and 59% for EGPM model for 500 ppm inlet concentration at 70 mm weir height and mass velocity of solids (141.5 x  $10^{-2}$  kg/m<sup>2</sup> ·s). It may be seen that the experimental value is 4.83% more than the theoretical value for EGPF model and 10.16% more for EGPM model. The percentage of sulfur dioxide is 52% for EGPF model and 49% for EGPM model for same inlet concentration at 70 mm weir height mass velocity of solids  $(71.0 \times 10^{-2})$  $kg/m^2$  ·s). It may be seen that the experimental value is 5.76% more than the theoretical value for EGPF model and 12.24% more for EGPM model. The trend indicates that decreasing in the mass velocity of solids decreases the removal efficiency, as the probability of collision between gas and solid particles decreases due to decrease in solids holdup.



Fig. 6. 3. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at  $h_W = 0.07$  m and  $G_S = 141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 6. 4. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at  $h_W = 0.07$  m and  $G_S = 71.0 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 

Figs. 6.5 and 6.6 represent the effect of inlet sulfur dioxide concentration on the percentage removal of SO<sub>2</sub> at a particular mass velocity of gas  $(31.2 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s})$ . Similar trends have been emerged. The percentage removal of SO<sub>2</sub> is also higher for EGPF model. The percentage removal efficiency of sulfur dioxide is 59% for EGPF model and 56% for EGPM model for 500 ppm inlet concentration at 30 mm weir height and mass velocity of solids  $(141.5 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s})$ . It may be seen that the experimental value is 4.83% more than the theoretical value for EGPF model and 10.71% more for EGPM model. The percentage of sulfur dioxide is 49% for EGPF model and 47% for EGPM model for same inlet concentration at 30 mm weir height mass velocity of solids  $(71.0 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s})$ . It may be seen that the experimental value matches with theoretical value for EGPF model and 14.16% less from EGPM model. It indicates that decreasing in the weir height decreases the removal efficiency, as the solids holdup in the bed decreases.


Fig. 6. 5. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at  $h_W = 0.03$  m and  $G_S = 141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 



Fig. 6. 6. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at  $h_W = 0.03$  m and  $G_S = 71.0 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ 

## Effect of mass velocity of gas on sulfur dioxide removal efficiency

The percentage removal of SO<sub>2</sub> at different gas flow rate and different inlet SO<sub>2</sub> loading has been plotted in Fig. 6.7 for both models. It may be seen that a lower gas velocity of  $31.2 \times 10^{-2} \text{ kg/m}^2$  ·s at mass velocity of solids of  $141.5 \times 10^{-3} \text{ kg/m}^2$  ·s has higher sulfur dioxide removal efficiency for both the models which is 59% for EGPM model and 62% for EGPF model for 500 ppm inlet concentration and 47% for EGPM model and 50% for EGPF model for 1500 ppm inlet concentration at weir height 70 mm. It may be seen that the experimental value is 6.0% more than the theoretical value for EGPF model and 12.76% more for EGPM model for inlet concentration 1500 ppm. The slight difference in removal efficiency between the predictions of two models tends to indicate that both model predicted well the overall removal efficiency, but with a sight advantage for the EGPF model. Increasing in inlet sulfur dioxide concentration and mass velocity of gas decreased the removal efficiency.



Fig. 6.7. Effect of superficial mass velocity of gas on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.07$  m and Gs = 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s (A) Inlet concentration = 500 ppm (B) Inlet concentration = 1500 ppm

Fig. 6.8 presents the effect of superficial mass velocity of gas on percentage removal of SO<sub>2</sub> at mass velocity of solids of 71.0 x  $10^{-3}$  kg/m<sup>2</sup> ·s and weir height 30 mm. Similar trends are also emerged for weir height 30 mm. For the given simulation, the sulfur dioxide removal efficiencies for EGPM and EGPF model are 59% and 62% for 500 ppm inlet concentration and 47% for EGPM model and 50% for EGPF model for 1500 ppm inlet concentration. It may be seen that the experimental value is 6.0% more than the theoretical value for EGPF model and 12.76% more for EGPM model for inlet concentration 1500 ppm and the EGPF value was much closer to the experimental value indicting valid assumption of plug flow for the emulsion phase compared to perfectly mixed condition.



Fig. 6.8. Effect of superficial mass velocity of gas on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.03$  m and Gs = 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s (A) Inlet concentration = 500 ppm (B) Inlet concentration = 1500 ppm

## Effect of weir height on percentage removal efficiency of $SO_2$

Fig. 6.9 represents the effect of weir height on the percentage removal of SO<sub>2</sub> at a particular mass velocity of gas  $(31.2 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s})$  and mass velocity of solids  $(141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s})$ . For this set of prediction, temperature was set to 310-311 K. It may be observed that the removal efficiency increases as the weir height increases. The percentage removal of SO<sub>2</sub> is higher for EGPF model compared to EGPM model. The percentage removal of sulfur dioxide is maximum for 70 mm weir height and minimum at 30 mm weir height for both the model. It may be seen that at weir height 50 mm and for 1000 ppm inlet concentration, the minimum sulfur dioxide removal efficiency is 49% for EGPM model and 52% for EGPF model and maximum sulfur dioxide removal efficiency is 54% for EGPM model and 56% for EGPF model. The trend indicates that decreasing in the weir height decreases the removal efficiency due to decrease in solids holdup.



Fig. 6. 9. Effect of superficial mass velocity of gas on percentage removal efficiency of SO<sub>2</sub> at  $h_w = 0.07$  m and Gs = 141.5 x 10<sup>-3</sup> kg/m<sup>2</sup> ·s (A) EGPF (B) EGPM

# Effect of temperature on percentage removal efficiency of sulfur dioxide

Fig. 6.10 describes the effect of temperature on sulfur dioxide removal efficiency for both models at 70 mm weir height. Since the reaction rate depends on temperature and overall desulphurization rate increased with increase in temperature, the removal efficiency was around 98-99% for both models at 120 °C for 500 ppm. The difference between the predictions of two models was very less as compared to difference at higher concentration. As the temperature decreased, the removal efficiency decreased at very faster rate.



Fig. 6. 10. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at different temperature

## 6.3 Stage wise percentage removal efficiency of sulfur dioxide

Fig. 6.11 indicates the comparison on percentage removal efficiency of  $SO_2$  between the experimental results and predicted results at different stages at a particular solid flow rate, gas flow rate and weir height. It may be seen that at the first stage, the maximum percentage of the inlet sulfur dioxide was absorbed and reacted at the surface of the calcium sorbent. As a result, the removal efficiency was higher than

other two stages. At the second stage, formation a product layer (CaSO<sub>3</sub>  $.0.5H_2O$ ) gradually at the surface of the sorbent decreased the removal efficiency, which was similar to that observed by Ho and Shih (1992) in a fixed bed.

The slight difference observed between the predictions of the two models tends to indicate that the gas interchange between the bubble and the emulsion phase is not the limiting step on this process. This is probably a consequence of rather narrow bed height.



Fig. 6. 11. Effect of inlet SO<sub>2</sub> concentration on percentage removal efficiency of SO<sub>2</sub> at different stages at T=38 <sup>o</sup>C

#### Comparison of experiment and theoretical efficiency

Fig. 6.12 shows a comparison between the tendency of the EGPM and EGPF model. The EGPF model predicted well as in close agreement with experimental data. The slight difference observed between the predictions of the two models tends to indicate that the gas interchange between the bubble and the emulsion phase is not the limiting step on this process. This is probably a consequence of rather narrow bed height.



Fig. 6. 12. Comparison of theoretical efficiency with experimental efficiency

#### 6.4 Summary of findings

A model based on assumption of Davison and Harrison (1963) with suitable modification has been developed for simulating the operation of counter-current multi-stage fluidized bed reactor. It is observed that the removal efficiency decreases as the inlet sulfur dioxide concentration increases. The percentage removal of SO<sub>2</sub> is higher for EGPF model compared to EGPM model in all cases and both the model values are compared with experimental values. It is observed that the experimental value is 5.76% more than the theoretical value for EGPF model and 12.24% more for EGPM model for 500 ppm inlet concentration at 70 mm weir height and mass velocity of solids and gas of 141.5 x  $10^{-3}$  kg/m<sup>2</sup> ·s and 31.2 x  $10^{-2}$  kg/m<sup>2</sup> ·s respectively. At same operating conditions with weir height 30 mm, the experimental value is 4.83% more than the theoretical value for EGPF model and 10.71% more for EGPM model. It indicates that decreasing in the weir height

decreases the removal efficiency, as the solids holdup in the bed decreases. The percentage removal of  $SO_2$  at different gas flow rate and different inlet  $SO_2$  loading is also calculated for both models. It is observed that a lower gas velocity at a particular solid velocity has higher sulfur dioxide removal efficiency for both the models which is 59% for EGPM model and 62% for EGPF model for 500 ppm inlet concentration and 47% for EGPM model and 50% for EGPF model for 1500 ppm inlet concentration at weir height 70 mm.

The effect of weir height on the percentage removal of  $SO_2$  is also calculated at a particular mass velocity of gas  $(31.2 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s})$  and mass velocity of solids  $(141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s})$ . It is observed that the removal efficiency increases as the weir height increases. At weir height 50 mm and 1000 ppm inlet concentration, the maximum sulfur dioxide removal efficiency is 54% for EGPM model and 56% for EGPF model.

The effect of temperature on sulfur dioxide removal efficiency for both models is calculated at 70 mm weir height. The removal efficiency is around 98-99% for both models at 120  $^{O}$ C for 500 ppm. Based on prediction, it can be showed that with increase in the temperature increases sulfur dioxide removal efficiency. It is found that the maximum SO<sub>2</sub> is removed in first stage of the reactor in comparison to second and third stage.

The removal efficiency of  $SO_2$  is found to be a strong function of inlet  $SO_2$  loading, mass velocity of gas and solids and weir height. The theoretical  $SO_2$  removal efficiency shows a good agreement with experimental results. The assumptions of plug flow of the gas percolating through the emulsion phase leads to slightly better predications than the assumption of perfect mixing of the emulsion phase. The slight difference in removal efficiency between the predictions of two models tends to indicate that both model predict well the overall removal efficiency, but with a sight advantage for the EGPF model.

Even though the some of conclusions are specific to the study, the model may be considered general enough to be used for predicting the performance of a countercurrent multistage fluidized bed reactor for any gas-solid treatment.

## **CHAPTER 7**

## CONCLUSIONS AND FUTURE SCOPE OF STUDY

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# Conclusions and future scope of study

#### 7.0 Introduction

In the present investigation; a multistage fluidized bed reactor have been developed, designed and fabricated to remove SO<sub>2</sub> from synthetic air-SO<sub>2</sub> mixture. It is necessary to know about the energy spent in the reactor by the carrier gas; hence the hydrodynamic studies of the reactor were carried out for the operating variables of mass velocity of gas ranging from  $31.27 \times 10^{-2}$  to  $56.40 \times 10^{-2}$  kg/m<sup>2</sup>·s and mass velocity of solids from  $35.4 \times 10^{-3}$  to  $141.50 \times 10^{-3}$  kg/m<sup>2</sup>·s at different weir heights. The performance study of the reactor for SO<sub>2</sub> removal was carried out for the same set of operating variables along with different inlet concentration of SO<sub>2</sub> from 500 to 1500 ppm. Mathematical modeling of the reactor for sulfur dioxide removal was also derived and simulated and the results were discussed.

#### 7.1 Conclusions

Conclusions were drawn based on the results obtained from both experimental and theoretical analysis and listed in detail in the following sections.

#### 7.1.1 Hydrodynamic studies for single particle system

Chapter 3 deals with hydrodynamic studies of three stage fluidized bed reactor in order to find out the stable operating range of the reactor in respect of mass velocity of gas and solids, mechanism of gas-solid contacting in the two-phase system, the overall pressure drop and holdup under different flow rates of the phases.

## Regime of fluidization and study on stable operation

At low mass velocity of gas, as the solids were let into the fluidized bed, solids filled the downcomer and piled up over the distributor plate of the top stage just below the downcomer. As the mass velocity of gas was increased in small increments over minimum fluidization velocity, the solids dispersed and started distributing in the top stage under fluidized condition. The solids were then transferred from stage to stage fluidizing in each stage. For a particular mass velocity of solids, the operation of multistage fluidized bed reactor was stable within a range of mass velocity of gas called as "lower operating mass velocity of gas" and "upper operating mass velocity of gas". The lower operating mass velocity of gas increased with increase in particle size and particle density. It also increased with increase in mass velocity of solids. The upper operating velocity increased also with increase in particle size and particle density, but it decreased with increase in mass velocity of solids. This indicated that the range of stable operation of multistage fluidized bed reactor with downcomers decreases with increase in mass velocity of solids, an observation in agreement with Kannan et al. (1994). The regime in each bed was standard bubbling regime whereas solids regime in each downcomer was from slip-stick flow to moving bed flow [Fan et al., 2007].

Once the stable operating range for different particles was determined, experiments were conducted on the reactor to measure the pressure drop at mass velocity of gas and solids ranging from  $31.2 \times 10^{-2}$  to  $56.4 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s}$  and  $35.4 \times 10^{-3}$  to  $141.5 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$  respectively with three weir heights ( $30 \times 10^{-3} \text{ m}$ ,  $50 \times 10^{-3} \text{ m}$  and  $70 \times 10^{-3} \text{ m}$ ). The solids taken for investigation were hydrated lime ( $426 \text{ }\mu\text{m}$ ), sand ( $426 \text{ }\mu\text{m}$  and  $600 \text{ }\mu\text{m}$ ). Pressure differences ( $\Delta P$ ) for each gas-solid flow rate inside the reactor were measured by the difference in the head ( $\Delta$ h) of carbon tetrachloride in the limbs of the U-tube manometers.

#### Pressure drop and solids holdup characteristics

Pressure drop studies were conducted at stable range for two-phase systems. While operating the system with solids, it was observed that all the stages of the reactor were identical in their operation as well as performance. Pressure drops across each stage and across the entire column were studied. In view of identical performance, the gas pressure drop due to solids across each stage was obtained from the difference between the pressure drop with and without solids.

The pressure-drops due to solids (lime: 426  $\mu$ m) were measured across each stage varying the gas to solid flow rates and weir heights. The pressure drop due to solids,  $\Delta$ Ps decreased with increase in the mass velocity of gas and increased with increase in the mass velocity of solids. Similar observations were reported by Pillay and Varma (1983) and Krishnaiah and Verma (1982). The minimum pressure drop occurring in each stage at maximum mass velocity of gas of 56.4 x 10<sup>-2</sup> kg/m<sup>2</sup>·s corresponding to minimum mass velocity of solids of 35.4 x 10<sup>-3</sup> kg/m<sup>2</sup>·s were 57.1,

103.3 and 143.5 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir heights respectively. The maximum pressure drops occurring in each stage at minimum mass velocity of gas of  $31.2 \times 10^{-2} \text{ kg/m}^2$  s corresponding to maximum mass velocity of solids of 141.5 x  $10^{-3} \text{ kg/m}^2$  s were 98.4, 139.6 and 185.1 N/m<sup>2</sup> at 0.03, 0.05 and 0.07 m weir height respectively. It appears that an increase in mass velocity of gas increases the particle-particle and particle-gas interaction leading to increase in particles outflow, resulting in decrease in the solids concentration in bed and hence reduces the pressure drop across the stage. [Mohanty and Meikap, 2008d].

Following same experimental procedure, the gas pressure-drops due to sand particles (426  $\mu$ m and 600  $\mu$ m) were measured in each stage varying the mass velocity of gas and solids and weir heights. Similar trends were also observed. Overall conclusions were that an increase in the particle diameter and particle density increased the frictional resistance resulting in increase in pressure drop [Krishnaiah and Verma, 1982; Papadatos et al., 1975; Mohanty and Meikap, 2008d].

Theoretically friction factor could be calculated from the following equation:

$$f = \frac{\Delta p_s}{0.5\rho_g u_g^2} \cdot \frac{d_p}{h_w}$$
(7.1)

Next, an attempt was made to correlate the friction factor with system variables and the following correlations was developed based on Buckingham's  $\pi$  theorem and multiple linear regression analysis. The functional relationship between friction factor and the dimensionless groups is as follows;

$$f = 9.0 \left[ \frac{u_s}{u_g} \right]^{0.24} \left[ \frac{gd_p}{u_g^2} \right]^{1.12} \left[ \frac{d_p}{h_w} \right]^{0.2} \left[ \frac{\rho_s - \rho_g}{\rho_s} \right]$$
(7.2)

This empirical correlation (Eqn.7.2) satisfies the experimental data of the present system satisfactorily [Mohanty and Meikap, 2008d].

It was found experimentally that the solids hold-up and pressure drop were interdependent. A correlation for solids holdup at each stage was developed, which is given below;

$$\frac{W}{Ah_{w}\rho_{s}} = 5.0 \left[\frac{u_{s}}{u_{g}}\right]^{0.24} \left[\frac{gd_{p}}{u_{g}^{2}}\right]^{0.12} \left[\frac{d_{p}}{h_{w}}\right]^{0.2}$$
(7.3)

This empirical correlation (Eqn.7.3) agrees well with the experimental data of the present system satisfactorily as reported by Mohanty and Meikap (2008b).

The mean residence time of solids,  $\tau$ , was defined as the ratio of solids holdup in the stage (W) to mass velocity of solids, m (=AGs), which was satisfactorily correlated by defining a modified dimensionless residence time,  $\tau$ .

$$\tau_{e} = \frac{\tau \gamma}{d_{p} h_{w}}$$
(7.4)

$$\tau_c = (1 - \varepsilon) \frac{\gamma}{u_s d_p} \tag{7.5}$$

A correlation was developed to determine the solids height in the downcomer, which agreed with the experimental values satisfactorily;

$$h_{S} = \frac{\rho_{g} u_{g}^{2}}{4\rho_{s} (1 - \varepsilon_{nf})} \left[ \frac{fh_{w}}{d_{p}} + \frac{1}{FC_{d}^{2}} \right]$$
(7.6)

## 7.1.2 Pressure drop studies of mixed-particle system

Experiments were carried out with mixed-particle system i.e. a mixture of dolomite (25%) and lime (75%) particles of size (426  $\mu$ m) and a mixture of dolomite (50%) and lime (50%) particles of size (426  $\mu$ m). In both the sets, variation of pressure drop with gas velocity was studied with different solids flow rates varying the weir heights.

#### (a) For 25% dolomite and 75% lime

The effect of gas velocity on pressure drop at different weir heights was studied. Pressure drop decreased with increase in mass velocity of gas at a constant mass velocity of solids due to less resistance of solid with increased gas flow rate as reported by Mohanty et al. (2008a).

(b) For 50% dolomite and 50% lime

The pressure drop trend was similar to that of previous mixture of solids. As the dolomite proportion increased in the mixture of solids the pressure drop also increased because dolomite is denser than lime and offered more resistance to gas flow as reported by Mohanty et al. (2008a).

## 7.1.3 Desulphurization by multi-stage fluidized bed reactor

The hydrated lime was used as adsorbent for the desulphurization. The physical characteristics such as Brunauer–Emmett–Teller (BET) specific surface area were measured along with chemical properties by thermogravimetric analyzer (TGA).

The multi-stage fluidized bed reactor, which was used for hydrodynamic studies, was also used here with some modifications in order to supply the synthetic air-SO<sub>2</sub> mixture in composition similar to that of the flue gas of sulfuric acid plants and copper smelters, to the system. The performance of the reactor for removal of SO<sub>2</sub> gas was carried out with different gas–solid flow rates and two weir heights. The range of operating parameters are: (i) Ambient air temperature,  $T_{ambient} = 308 \pm 3$  K, (ii) Mass velocity of gas,  $G_a = 31.2 \times 10^{-2}$  to  $56.4 \times 10^{-2}$  kg/m<sup>2</sup>·s, (iii) Mass flow rate of solids,  $G_S = 35.4 \times 10^{-3}$  to  $141.5 \times 10^{-3}$  kg/m<sup>2</sup>·s (iv) Inlet gas concentration,  $C_i = 500$  ppm to 1500 ppm. The study was carried out at ambient temperature and a pressure of 1 atmosphere. Under steady state operating conditions, the SO<sub>2</sub> gas samples were collected at each point at flow rate of 1.0 LPM with the help of impinges and aspirator bottles. The gas samples were analyzed for sulfur dioxide by the "Tetrachloro-Mercurate method" [IS: 5182(Part-VI]. Percentage removal of SO<sub>2</sub> has been calculated for each experimental run by the formula [Levenspiel, 1962].

$$\eta_{SO_2} = \frac{SO_{2Inlet} - SO_{2Outlet}}{SO_{2Inlet}} x100$$
(7.7)

(7.8)

Likewise for stage i,  $\eta_{SO2} = (C_{i+1}-C_i) / C_{i+1}$ 

Where  $C_i$  and  $C_{i+1}$  are outlet and inlet sulfur dioxide concentrations in gas.

The conclusion arrived are summarized below.

## Effect of gas flow rate at different SO2 loading on the percentage removal of SO2

The percentage removal efficiency of SO<sub>2</sub> ( $\eta_{SO2}$  %) was measured by varying the mass velocity of gas and solids, inlet SO<sub>2</sub> loading and weir heights. It was observed that lowering the mass velocity of gas at a particular mass velocity of solids had higher sulfur dioxide removal efficiency of 65% at 70 mm weir height and 62% at 30 mm weir height. This is due to the fact that lowering the mass velocity of gas increases the gas residence time resulting in higher removal of gas. Same trend was reported by Jiang et al. (1995) in a circulating fluidized bed adsorber and Mohanty et al. (2008c) in a multistage fluidized bed reactor. It was also found that the increasing inlet sulfur dioxide concentration decreased the removal efficiency, which may be due to an increase in sulphite concentration on the pores of lime particles, thus resulting in decrease of the adsorbent activity.

## Stage wise percentage removal of sulfur dioxide

It was observed that at the first stage, the solid reactant was fresh for which maximum percentage of the inlet sulfur dioxide was adsorbed and reacted at the surface of the calcium oxide sorbent. As a result, the removal efficiency was higher than other two stages. At the second stage, formation of product layer (CaSO<sub>3</sub> .0.5 H<sub>2</sub>O) at the surface of the sorbent perhaps increased the diffusion resistance of SO<sub>2</sub> from the emulsion phase to the inner untreated calcium sorbent, thus decreasing the removal efficiency. The maximum removal of sulfur dioxide at first stage was found to be 40% at mass velocity of gas of  $31.2 \times 10^{-2} \text{ kg/m}^2 \cdot \text{s}$ , mass velocity of solids of  $35.4 \times 10^{-3} \text{ kg/m}^2 \cdot \text{s}$ , inlet gas concentration of 500 ppm and weir height of 70 mm.

# 7.1.4 Development of model for removal of sulfur dioxide in a multi-stage fluidized bed reactor

A model based on gas and solid phase material balance was applied with modification to study the removal of sulfur dioxide (SO<sub>2</sub>) in the multistage fluidized bed reactor [Davidson and Harrison, 1963; Hymore and Laguerie, 1984]. Sulphur dioxide concentrations in the exit gas stream from the i<sup>th</sup> stage was calculated by using emulsion gas perfectly mixed (EGPM) model and emulsion gas in plug flow (EGPF) model:

(a) Sulphur dioxide concentration in the exit gas stream from the  $i^{th}$  stage: The rate of SO<sub>2</sub> disappearance in the  $i^{th}$  stage can be expressed as:

$$r_i = k C_{ri} C_{pi} \tag{7.9}$$

For the concentration of the gas stream leaving the ith stage:  $C_{i} = \beta C_{bi}(H) + (1 - \beta)C_{pi}$ (7.10)

EGPM Model:

$$C_{\mu}(H) = C_{\mu} + (C_{\mu} - C_{\mu})e^{-X}$$
(7.11)

$$C_{pi} = \frac{C_{i+1} (1 - \beta) e^{-X}}{\left(1 - \beta e^{-X} + \frac{H_{mf}}{U} r_i\right)}$$
(7.12)

EFPF Model:

$$C_{pi} = \left(\frac{C_{i+1}}{m_{1i} - m_{2i}}\right) \left[m_{1i} \left(1 + \frac{h_w}{X} m_{2i}\right) \exp(m_{2i} h_w) - m_{2i} \left(1 + \frac{h_w}{X} m_{1i}\right) \exp(m_{1i} h_w)\right]$$
(7.13)

$$C_{bi} = \left(\frac{C_{i+1}}{m_{1i} - m_{2i}}\right) \left[m_{1i} \exp\left(m_{2i}h_{W}\right) - m_{1i} \exp\left(m_{1i}h_{W}\right)\right]$$
(7.14)

To solve the model equations, a simulation program was developed in Matlab and the results were presented. The percentage removal efficiency of  $SO_2$  ( $\eta so_2$  %) at different inlet  $SO_2$  loading, mass velocity of gas and solids were predicted using EGPM and EFPF models. Besides these parameters, the effect of temperature was also studied. It was observed that at lower mass velocity of gas, the EGPF model predicted data well with that of experimental findings. The slight difference observed between the predictions of the two models indicated that the gas interchange between the bubble and the emulsion phase may not be the limiting step in the adsorption process.

#### 7.2 Conclusions

In the present investigation; a three-stage counter current fluidized bed reactor has been fabricated and operated in bubbling fluidization regime to study the hydrodynamic characteristics of the reactor for single and mixed particles system and removal efficiency of sulfur dioxide from synthetic SO<sub>2</sub>-air mixture using hydrated lime particles. The operating limits of mass velocity of gas and solids are obtained for the stable operation of the fluidized bed reactor. The pressure-drop across each stage increases with increasing in weir height or mass velocity of solids and decreases with increase in superficial mass velocity of gas and the maximum pressure drop for lime particle at each stage is found to be 185 N/m<sup>2</sup> at 70 mm weir height. The maximum pressure drops for mixed particles ( 50% lime and 50% dolomite) occurring in the column at low mass velocity of gas (31.2 x  $10^{-2}$  kg/m<sup>2</sup>·s) corresponding to maximum solid flow rate (84.7 x 10<sup>-3</sup> kg/m<sup>2</sup>·s) are 143.8, 151.6, and 178.8 N/m<sup>2</sup> at 0.04, 0.05, and 0.06 m weir heights respectively. Although maximum pressure drops are obtained for higher dolomite ratio mixture, no choking of material inside the downcomer is observed. The hydrodynamic data presented in this study assume significance from the perspective of design and stable operation of staged fluidized bed reactors.

Experiments have been conducted to ascertain the  $SO_2$  removal efficiency in the multistage fluidized bed reactor at different mass velocity of solids and gas. It is observed that at the first stage, the solid reactant is fresh for which maximum percentage of the inlet sulfur dioxide is absorbed. The removal efficiency is higher in first stage (40%) and lowest in last stage (19%) corresponding to overall removal efficiency (65%) at gas flow rate (31.2 x  $10^{-2}$  kg/m<sup>2</sup>·s), solids flow rate (141.5 x  $10^{-3}$  kg/m<sup>2</sup>·s) and weir height (0.07 m). A model is used for simulating the removal of SO<sub>2</sub> in the reactor. The assumption of plug flow of the gas percolating through the emulsion phase leads to slightly better predication than the assumption of perfect mixing of the gas in emulsion phase. The scope of the future work is presented in the light of the results and conclusions drawn above.

#### 7.3 Future scope and recommendations of the work

The present investigation on sorption of  $SO_2$  gas has been done only for few inlet concentrations of  $SO_2$  loading rates in the reactor, in reality the concentrations fall in a wide range. More over there are plenty of air pollutants emitted from various industries, thus sorption potentiality of the reactor has to be studied for wide range of pollutants in both individual and synergetic forms.

However, due to paucity of time, the following could not be completed and may be taken up as future work.

- The RTD of solids and gas may be carried out for clear understanding of the sorption process.
- Performance studies of the reactor for scrubbing of other gaseous pollutants individually and in a mixture of NOx, CO<sub>2</sub>, CO etc. in different concentrations at high temperature.
- Operation of the system with recycles of solids to optimize the sorbent utilization.
- CFD simulation of reactor performance may be undertaken to predict the removal efficiency of individual and combined pollutants.

• Economic viability of the process with pilot plant testing in real plant.

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#### List of publications: (A) Journal papers published

- <u>C. R. Mohantv.</u> S. Adapala, B. C. Meikap, Hydrodynamics of a multistage countercurrent fluidized bed reactor with downcomer for limedolomite mixed particle system, Industrial & Engineering Chemistry Research, 47(18), 6917–6924, 2008.
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- 3. <u>C. R. Mohanty</u>, S. Adapala, B. C. Meikap, Holdup characteristics of a novel gas-solid multistage fluidized bed reactor for control of hazardous gaseous pollutants, **Chemical Engineering Journal**, 148(1), 2009
- C. R. Mohanty, B. C. Meikap, Removal of hazardous gaseous pollutants from industrial flue gases by a novel multistage fluidized bed desulfurizer, Journal of Hazardous Materials, In press, 2008.
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#### (B) Papers under review

1. <u>C. R. Mohanty</u>, B. C. Meikap, Theoretical and experimental studies on stable operating range of a multistage fluidized bed reactor with downcomer, *Chemical Engineering and Processing: Process Intensification.* 

#### (C) Conference/Symposia/Workshop/Short courses etc.

1. <u>C. R. Mohanty</u>, B. C. Meikap, 2008. Characterization of a multi-stage counter-current fluidized bed reactor for abatement of hazardous industrial gaseous effluents, Proceedings of AICTE short course on "Advanced fluidization techniques for waste management and energy production", NIT Rourkela.

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