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Hydrodynamic of a co-current gas liquid upflow in a moving packed bed reactor with porous catalysts

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HYDRODYNAMIC OF A CO-CURRENT GAS LIQUID UPFLOW IN A MOVING PACKED BED REACTOR WITH POROUS CATALYSTS

By

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This thesis consists of the following two papers, formatted in the style utilized by Missouri University of Science and Technology. Paper I, comprising pages 9 through 55, is submitted to the Energy & Fuels journal, under the tittle ‘‘Liquid Holdup Studies In A Co-current Gas Liquid Upflow Moving Packed Bed Reactor With Porous Catalyst Using Gamma Ray Densitometry’’. Paper II, comprising pages 56 through 94, is submitted to the Chemical Engineering Science journal under the title ‘‘Identification Of Flow Regime In a Co-current Gas – Liquid Upflow Moving Packed Beds Reactor Using Gamma Ray Densitometry’’.

ABSTRACT

In this study, the hydrodynamic, i.e. flow regime identification, line average liquid holdup, and the internal liquid holdup of a co-current moving packed bed reactor were studied. For the sake of hydrodynamics study, moving bed reactor is investigated as two – phase upflow packed bed reactor. Scaled down configuration was used to simulate of the moving bed reactors utilized in the industrial process. First, line average liquid holdup is measured with different geometrical configurations covering empty, dry, wet, and packed column under flowrates operation conditions. A new methodology has been developed to determine the line average liquid holdup for a porous catalyst. Second, flow regime is identified by variation of superficial gas velocity at constant liquid superficial velocity. The experiments were carried out in an 11 - inch inner diameter Plexiglas column using the air-water system, at superficial gas velocities in the range of 0.6 to 7.7 cm/s and at a constant liquid superficial velocity of 0.017 cm/s. Gamma ray densitometry (GRD) technique was used to obtain the line average liquid holdup and to identify the flow regime at different axial and radial positions along the column. The obtained results showed that the flow regimes are bubble flow and pulse flow regimes with a transition flow in-between under the operation conditions used. The result showed that the liquid holdup decreased as the superficial gas velocity increased. It was also found that the liquid holdup radial distribution was not uniform. These kinds of information are essential to improve the performance of the reactor.

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1. INTRODUCTION

Multiphase flow reactors and multiphase flow are encountered widely in industrial processes where more than one phase (gas-liquid, and solid) interact in a complex manner. Moving packed bed reactors belong to a general class of catalytic multiphase flow reactors, which are consisting of a combination of gas, liquid, and solid phases. Moving bed reactors exist in hydrotreating industries, such as hydrodesulfurization, hydrodenitrogenation, and hydrodemetallization, etc. They are typically vertical reactors with gas and liquid flow upward through catalyst particles bed that periodically move downward. These reactors have the distinctive design of its bottom that support the catalyst and facilitated their removal periodically while fresh catalyst are added at the top of the bed. Hence, catalysts of the moving bed reactor are supported by a cone-like shape bottom to enable withdrawn the deactivated catalysts. The catalysts move downward in a counter current mode with the two phase flow of the gas and liquid. The catalyst removal would occur at a rate of 2 to 8% per week depending on the feed metals content, and that is in small increments with no interruption to the process. The advantage of continuous regeneration processes in the moving – bed processes, is to avoid the process interruption (Liu et al., 2009; Reynolds et al., 2003). These reactors operate at the condition of incipient fluidization and hence the top level of the catalyst bed faced a slight expansion and fluidization, which reduces the pressure drop and avoids plugging of the system (Liu et al., 2009). The moving bed is usually used as a leading guard reactor in front of the conventionally hydrotreating fixed bed to prolong the operating cycle (Liu et al., 2009). The moving – bed configuration,

offers a relatively large catalyst migration time, in comparison with the liquid and gas phases mean residence time, and it can be considered as a pseudo-two-phase (gas-liquid) flow fixed bed (Iliuta and Larachi, 2013a) but with a configuration of the bottom of the catalysts support differs from the conventional fixed bed two-phase upflow reactors. Therefore, for the sake of hydrodynamics study, moving bed reactor is investigated as two – phase upflow packed bed reactor with the bottom design represents the upflow moving bed. The understanding of the hydrodynamic of these reactors is essential for proper design, scale-up, operation and performance. The knowledge of the flow regime, the liquid holdup and bed structure are indispensable in the understanding and analysis of the moving upflow packed bed reactors and their performance.

Three types of flow regimes are commonly observed in upflow packed beds. These flow regimes are bubble flow, pulse flow regime, and spray flow with water and air as liquid and gas phase (Raghavendra Rao et al., 2011; Varma et al., 1997). Flow regimes vary depending on operation conditions, fluid and packing properties, and flow rates. They are typically categorized in co-current upflow packed bed reactors into low interaction regime (bubble flow) and high interaction regime (pulse and spray flow regimes). Depending on the level of interaction between phases, therefore, each regime has different hydrodynamic characteristics which affect the rate of the mass and heat transfer, pressure drop, and liquid holdup. Bubble flow regime occurs at low gas and liquid flowrates, which is characterized by small bubbles flow as a dispersed gas in a continuous liquid within the void of the bed. The buoyancy force leads to rise the bubbles in the upward flowing liquid in the bubbly flow regime where the bed particles are fully wetted by the continuous liquid phase (Guo and Al-Dahhan, 2005). This flow regime refers as a low interaction regime, as

there is a little interaction between the liquid and gas phases and their friction with the solid particles. Pulse flow regime occurs at moderate to high gas and liquid flow rates and has a very interesting hydrodynamic behavior. In this regime, the flow is characterized by an alternating gas rich region followed by liquid rich region. It occurs in packed beds due to increasing the diameter of the gas bubbles, which may not bridge the entire column cross-section (Raghavendra Rao et al., 2011). Pulse flow regime refers as a high interaction regime, in which high degree of mixing and interaction between the phases and with the solid particles enhance overall heat and mass transport while reducing axial dispersion. This makes it a potentially attractive mode of operation for industrial processes. When further increasing in gas superficial velocity and at low liquid superficial velocity, spray flow regime occurs. This flow regime is characterized by liquid droplets entrained in the packed bed by the higher gas flow rate. Another important hydrodynamic parameter in catalytic multiphase reactor is the liquid holdup, which is considered as one of the significant design and operating variables. In three phase contacting systems, the interactions between gas – liquid – solid is more complicated and hence, the liquid holdup is essential for achieving desirable of pressure drop, mass and heat transfer where the liquid phase serves to transport mass and heat to and from the catalyst bed particles.

The knowledge of liquid holdup is a key in the reactor design and model calculations of the reactor performances, and it's one of the important hydrodynamic parameters for the gas – liquid flow in upflow moving packed bed reactors. In a packed bed, the liquid holdup is affected by the gas superficial velocity more than the liquid velocity (Molga and Westerterp, 1997b). For exothermic reactions, higher liquid holdup and well distributed ensure a complete wetting efficiency and better temperature control

thus contributing to the prevention of hot spots formation and thermal instabilities. Two phase upflow packed bed reactor could be a satisfactory alternative to the classical trickle bed reactor for liquid limited reactions, because of increased liquid holdup to ensure a complete wetting efficiency of the catalytic where increase the efficiency of the solid contact lead to better heat transfer and higher overall mass transfer coefficient (Al-Dahhan and Duduković, 1996).

These hydrodynamic parameters in a co-current upflow packed bed reactors such as flow regimes and liquid holdup have been studied by many researchers (Bouteldja et al., 2013; Kumar et al., 2012; Moreira and Freire, 2003b; Murugesan and Sivakumar, 2002; Raghavendra Rao et al., 2011; Varma et al., 1997) for packed bed with a horizontal perforated distributor. While, there is no work has been done on moving packed bed configuration with a conical bottom. Therefore, in our work, an attempt has been made for the first time to identify the prevailing flow regimes and to measure the line average liquid holdup in this type of catalytic multiphase reactors using the newly developed non-invasive gamma ray densitometry technique.

2. MOTIVATION

The Recent trend toward improving the quality of light fuels, converting heavier oils into lighter and more valuable products efficiently while protecting the environment have become a challenge for refineries to come up with advanced refinery processes. The availability of heavy crude oil and residuum feed make the fixed bed catalyst system difficult to handle feedstock with more than 250 ppm level of metal contaminations. As an advanced residue hydroconversion, moving bed processing can deal with a heavier metal content feedstock up to 400 ppm (Liu et al., 2009). The best utilization of the hydroprocessing is by employing the moving bed reactor as a guard reactor before the conventional fixed bed reactor. Since the first commercial unit has been started in operation in the 1990's, there are five commercial units worldwide in operation (Liu et al., 2009). Some problems associated with the operation of these reactors are maldistribution, hot spot, and reduced expected conversion. To overcome these challenges, detailed studies to enhance the understanding of the hydrodynamics parameters that characterized this reactor are still required, and unfortunately, they lack in the open literature. Although research has been conducted on hydrodynamic of upflow packed bed reactors with a horizontally bottom plate distributor, but to the best of the author's knowledge, there is no study to evaluate hydrodynamic parameters in the open literature has accounted for the presence of a cone – like shape bottom in these reactors which represent the design of the upflow moving bed reactor. The knowledge of the flow regime and the liquid holdup are of great importance for the proper design scale-up, operation and performance of the upflow moving packed bed reactor as mentioned earlier.

In order to accomplish an improved understanding of the hydrodynamics of upflow moving packed bed reactors equipped with a cone – like shaped bottom, an investigation of the flow regime and line average liquid holdup by using newly developed advanced non-invasive gamma ray densitometry technique which are considered among the most important hydrodynamic parameters that govern the performance of packed beds reactors, is required. Investigations on these parameters in moving beds are currently not available in the open literature.

3. RESEARCH OBJECTIVES

The primary objective of this work is to advance the knowledge of the *Hydrodynamic of the Co-Current Upflow Moving Packed Bed Reactors with a Porous Catalyst*. To achieve this goal, extensive experimental work has been performed to identify and study the flow regime and to measure the line average liquid holdup in the two phase upflow moving packed bed with a cone shaped like bottom which is packed randomly with porous catalysts. The following objectives are set for this work as the main parameters to study:

1. Investigate the indentification of the flow regiemes under flow rates that represent the typical operating conditions using a scaled down two-phase upflow moving bed reactor. Our newly gamma ray densitometry has been implemented.
2. For the first time, we study and meausre the line averaged external and internal liquid holdups and the bed porosity. Along the bed diameter at selected heights using our newly developed gamma ray densitometry.

For the sake of the hydrodynamics study, a scaled down configuration of moving bed with two phase upflow has been used. The experiments have been conducted using an air-water system in a Plexiglas column with a diameter of 11 – inch and height of 30 – inch. The column packed randomly with a 3 mm diameter spherical catalysts with a total bed height of 24 – inch. The results and findings of this work will enhance the understanding of the hydrodynamics of moving packed bed reactors equipped with a cone – like shaped bottom, and it will facilitate the proper operation of these kinds of reactors.

In addition, the obtained data serve as valuable information for evaluation and validation of the objectives above have been achieved through the following two manuscripts:

1. Liquid holdup studies in a co-current gas liquid upflow moving pakced bed reactor with a porous catalyst using a new methodology for gamma ray densitometry.
2. Identification of flow regime in a co-current gas – liquid upflow moving packed bed reactor using statistical and chaos analyses.

PAPER

I. LIQUID HOLDUP STUDIES IN A CO-CURRENT GAS LIQUID UPFLOW MOVING PACKED BED REACTOR WITH POROUS CATALYST USING GAMMA RAY DENSITOMETRY

ABSTRACT

The upflow moving packed bed reactor are heavily used in hydrotreating. The problems associated with this reactor are maldistribution, hot spot, and reduced expected conversion, and found to be directly linked to liquid phase maldistribution. To overcome these challenges, detailed study to enhance the understanding of liquid phase hydrodynamics parameters that characterized this reactor is still required. The liquid holdup is one of the significant and important parameters in mass transfer and design of packed bed reactors, as it serves as heat and mass transport from and to the catalyst of the packed bed. In this work, the line-average liquid holdup was determined using Gamma-Ray Densitometry (GRD) in a scaled down lab scale upflow packed bed column. Gamma ray densitometry is a non-invasive radioactive technique that can be implemented to monitor the flow distribution even at industrial scale. There are no studies reported on the determination of the line average phase distribution for porous catalyst packed bed having conical bottom. In this study, a new methodology has been developed to determine the line average liquid holdup for the porous catalyst, line average void space of catalyst bed, and the line average internal porosity of catalyst. This study has been conducted on a Plexiglas column of 11-inch ID and 30-inch height, randomly packed with extrudate catalyst of 3 mm diameter till 24-inch height. GRD scanning is conducted at various axial and radial locations. The operation conditions for the study are at superficial liquid (water) at 0.017

cm/sec and varying superficial gas (air) velocity in the range of 0.6 – 7.7 cm/sec. The result showed decrease in liquid holdup with increasing in the superficial gas velocity, and the liquid holdup radial distribution was seen to be non-uniform. These kinds of information are essential to improve the performance of the reactor.

Keywords: Gas-liquid-solid reactor, Porous Catalyst, Upflow Moving Packed Bed Reactor, Line Average Liquid Holdup, Gamma Ray Densitometry.

1. INTRODUCTION

Improving the quality of light product and the increasing demand for heavy oil in compliance with strict environmental concerns have become a challenge facing leading petroleum industry. Catalytic hydroprocessing considers one of the most promising technologies for conversion of heavy oils into high-value products, and it has been extensively practiced nowadays in refineries worldwide (Liu et al., 2009). These aspects represent a motivation for developing the existing processes or designing specific advanced refining processes, related to hydroprocessing. Hydrotreatment (hydroprocessing) is a well know technology to remove undesirable components (sulfur, nitrogen, organometallic, etc) from hydrocarbon feed streams. Such hydroprocessing conditions are typically in the range of 212° F to 1200° F (100° to 650° C) at pressures from 20 to 300 atmospheres (Stangeland et al., 1991), at these operation conditions catalyst deactivate. In hydrotreatment process, the main issue is the catalyst life and its performance, because impurities can deposit on the catalyst resulting in rapid loss of its activity and deactivate it. Under high temperature and pressure, coke, poisoning, and sintering could cause agglomeration and hence maldistribution in which then the unit shutdown is unavoidable. Contaminating metals, such as nickel and vanadium, usually will be readily removed under hydrotreating conditions and will plate out on the surface and in the pores of the catalyst. The deposition of metals on the catalyst will result in a rapid loss of hydrogenation activity. However, hydrogenation activity is necessary for the removal of other contaminants, such as carbon residue, nitrogen, and sulfur, from the feedstock. Coke and various hydrocarbon products deposit on the particles and deactivate them as well. Various designs of residue hydrotreating reactors have been described in the literature for treating heavy feedstocks.

Commercial designs include a moving bed of catalyst (MBR), (U.S. Pat. No. 5,076,908), fixed bed reactor (FBR) (U.S. Pat. No. 5879642 A), ebullating catalyst bed (EBR), (U.S. Pat. Nos. 4,571,326 and 4,744,887). The fixed bed catalyst systems deal with middle distillates feed, but they cannot deal with every residuum feed that is available. Heavy feeds with highly metallic contaminations > 250 ppm, make the fixed bed catalytic hydrotreating system inefficient as run length become too short because of catalyst deactivation (Scheuerman et al., 1993). On the other hand, the high metals feeds which make fixed bed residuum desulfurization (RDS) units impractical are often the most economically attractive feeds because of their relatively lower price. To deal with drastic change in heavy petroleum feed properties, moving bed technology has been developed (Liu et al., 2009). In general, the moving bed reactors are having a conical bottom in which gas – liquid moves co-currently upwards and catalyst moves downwards periodically and spent catalyst are replaced using a conical bottom support. In this case, the flow of upward fluid with a slight bed expansion could avoid coking and plugging and reduce the pressure drop of the system to some extent (Liu et al., 2009; Reynolds et al., 2003). As the feed moves up through the catalyst and contaminants are retained on the catalyst, these particles become heavier and by periodically withdrawing they move downward through the reactor towards the entering products stream which is finally withdrawn at the bottom of the reactor. The removed catalysts can be reprocessed and injected alone or in combination with fresh catalyst at the top of the reactor. The advantages of using moving packed bed reactors are: it has represented a good methods for utilizing space within a hydroprocessing vessel, characterized by a slight back mixing for both the catalyst and the feedstock, providing a better quality of products and higher efficiency of the process than of an ebullated bed

reactor (Liu et al., 2009), the continuous replacement of spent catalyst without shutdown. The moving – bed configuration, offers a relatively large catalyst migration time, in comparison with the liquid mean residence time, and it can be considered as a pseudo two-phase (gas-liquid) upflow with fixed bed (Iliuta and Larachi, 2013a). Therefore, for the sake of the hydrodynamics study, the moving bed reactor can be investigated as two – phase upflow packed bed reactor. The liquid holdup in upflow moving packed bed reactor is one of the important design and operating hydrodynamic variables, and it measurement is essential to get a better understanding for the prediction of pressure drop, mass and heat transfer mechanisms since the liquid serves as a transport of mass and heat to and from the catalyst bed particles. Moreover, the upflow packed bed reactor gives advantages for liquid limited reaction (Chander et al., 2001). For exothermic reactions, higher liquid holdup and well distribution ensure a complete wetting efficiency and better temperature control thus contributing to the prevention of hot spots formation and thermal instabilities. Two-phase upflow packed bed reactor could be a satisfactory alternative to the classical trickle bed reactor for liquid limited reactions, because of increased liquid holdup to ensure a complete wetting efficiency thus increase the effectiveness of contact leading to better heat transfer and higher overall mass transfer coefficient (Al-Dahhan and Duduković, 1996). Total liquid holdup (ε_{Lt}) is defined as the ratio of the liquid volume in the bed to the total bed volume. Total liquid holdup can be divided into two types: external and internal liquid holdup, according to the type of catalyst used in the packed column in both upflow and downflow modes of operation (Al-Dahhan and Highfill, 1999). For nonporous particles, there is no internal liquid holdup. The ratio of the volume of the liquid occupying the void volume of the bed (void between particles) to the reactor volume is the external liquid

holdup (liquid contained outside porous particles). For porous particles, the internal liquid holdup is the ratio of the volume of the liquid held by capillary forces in the pores of porous catalysts to the reactor volume (liquid contained inside porous particles) (Al-Dahhan and Highfill, 1999). In packed bed, liquid saturation has been sometimes used instead of the liquid holdup in the description of liquid retention in the bed regarding the void volume or the bed volume. The volume of the liquid occupied in voidage between the catalytic particles to the void volume of the reactor is described as the external liquid saturation (Al-Dahhan and Highfill, 1999; de Klerk, 2003). The relationship between the external liquid saturation and external liquid holdup are interrelated as follows (Al-Dahhan and Highfill, 1999; Bensetiti et al., 1997; Jagadeesh-Babua et al., 2007).

$$\varepsilon_{L\,ext.} = \beta_{L\,ext.}\varepsilon \tag{1.1}$$

Where:

$\varepsilon_{L\,ext.}$: External liquid holdup, $\beta_{L\,ext.}$: External liquid saturation, and ε: porosity of the bed.

Experimental data on the overall liquid holdup or saturation can be obtained by various techniques such as for example drainage (Iliuta and Thyrion, 1997; Moreira et al., 2004; Urrutia et al., 1996), weighing (Kumar et al., 2012) and tracer methods (Cassanello et al., 1998; Guo and Al-Dahhan, 2004; Saroha and Khera, 2006; Thanos et al., 2003), Electric capacitance tomography (ECT) (Bouteldja et al., 2013; Hamidipour and Larachi, 2010). The liquid holdup measurements techniques can be divided into integral, semi – integral and local measurements methods (Al-Dahhan and Highfill, 1999). Integral methods provide liquid holdup information over the entire volume of the packed bed, these methods include the draining, weighting and tracer methods. Semi – integral measurement

methods provide liquid holdup information over a section or a line – integral of the packed bed, which include radiation methods (e.g. gamma Ray and X – Ray), that can be applied at many axial and radial positions in order to get a line averaged information. The local measurement methods provide a local liquid information are obtained by inserting a sensor (e.g. electromagnetic radiation) or a probe (e.g. optical fiber probe) inside the packed bed at different positions or using time averaged tomography (gamma ray tomography) or instantaneous tomography (X-ray and electrical capacitance resistive tomography). However, liquid draining and tracer methods can only give the average holdup for a whole packed column; it cannot offer any information on how the liquid is distributed in packings (Yin et al., 2002). In Fact, the liquid holdup can vary with spatial position and this information is very important for a better understanding of flow hydrodynamics and mass transport in packed columns. Since, in gas – liquid – solid conditions where the catalytic bed being dense and opaque, it is hard to implement instrumentation inside three phase systems. While the noninvasive methods such as advanced radioactive measurements techniques eliminate the alteration during the measurements (Hubers et al., 2005). These techniques can determine the flow distribution over the whole reactor section with a good spatial resolution and that are not too intrusive (Boyer et al., 2002). The radiation method is based on attenuation of the radiation beam as it passes through an absorption medium, where the liquid holdup can be obtained by Beer-Lambert's equation. Non-invasive techniques have become the tools of choice in pursuance of the detailed flow structures within porous media unlike the more traditional interfering probes inserted within flows (Hamidipour and Larachi, 2010).

Attempts have been made to study the liquid holdup in two phase co-current upflow packed bed reactor which are summarized as follow: (Bouteldja et al., 2013) studied the effect of inclination on the hydrodynamic of gas–liquid cocurrent upflow packed beds. They measured the Liquid saturation, bed pressure drop and gas–liquid segregation. They found at vertical position and constant liquid velocity the increasing in the gas velocity will lower the liquid saturation because the higher presence of gas phase. They observed from their results that bed inclination creates short circuits for the gas phase along the upper wall where it can flow in a segregated manner. (Kumar et al., 2012) studied the liquid holdup in upflow packed bed with two types of packing (randomly and structured packing) and they found that the liquid holdup in structured packing is 50% higher than the randomly packing catalysts. They observed that both total and dynamic liquid holdup in randomly packings decreased with increasing in gas flowrate, increased with liquid flowrates and viscosity of the liquid. They also observed the combine effect of bed porosity and size of the packing on both liquid holdups. They developed correlations for total and dynamic liquid holdup in both corrugated structured packings and the random packings as well. (Saroha and Khera, 2006) studied three hydrodynamic parameters of fixed beds with cocurrent upflow and downflow: two-phase pressure drop, total liquid holdup and axial dispersion and they compared the results for both modes of operation. They found that at low gas and liquid velocities the two phase pressure drop and liquid holdup for upflow is higher than the downflow. They also observed by increasing the flowrates these parameters became comparable for both the upflow and downflow. (Colli-Serrano and Midoux, 2000) measured the liquid holdup in two phase upflow by conductimetric probes using a salt tracer technique. They studied the influence the coalescence inhibition. They used nitrogen

as a gas and both water and aqueous solutions of pentanol (pOH) flow upwards to get their data. Their experiments performed in a packed bed that heated electrically through its wall. They obtained the heat transfer parameters by fitting a two-dimension model. They found that the heat transfer is strongly depend on the flow regime in the packed bed. Liquid holdup for (pOH solution) is lower than water as coalescing system. Their result leaded to a two experimental correlations for single and two phase flow. (Cassanello et al., 1998) studied the liquid saturation and back mixing in cocurrent upflow three phase fixed bed reactors using residence-time distributions (RTD). They tested two types of correlations for liquid saturation, one based on dimensionless number and the second one based on the drift flux concept. They found that the drift flux correlation is account with their experimental results. (Bensetiti et al., 1997) proposed a correlation for the prediction of external liquid saturation, their correlation relied upon combination of dimensional analysis and artificial neural networks that allowed identification of six most expressive dimensionless groups. (Iliuta and Thyrion, 1997) analyzed the hydrodynamic in packed bed operated with upflow and downflow using air/Newtonian and non-Newtonian fluid systems. They studied the liquid holdup (dynamic and residual liquid holdup) and found its value is strongly influenced by using non-Newtonian liquids. Their result for liquid holdup in two phase upflow and downflow are close to each other with highly viscous non-Newtonian liquids. However, to the best of our knowledge, there is study reported in the literature that studied liquid holdup in two phase upflow moving beds where the bottom configuration differs, with the present of the cone from the conventional two phase upflow packed bed reactors. Radiation techniques are commonly used in a wide range of applications as measuring methods since they are generally considered simple and non-

intrusive. Radiation methods include the use of fast neutron scattering and attenuation techniques as well as the use of gamma and X-ray attenuation techniques. Since the catalytic packed bed is opaque, some noninvasive visualization technique such as such as Digital Particle Image Velocimetry (DIPV) and Laser Doppler Anemometry (LDA), cannot measure the liquid/gas holdup distribution over its cross-section. Compared with other radiation techniques, the gamma ray technique is well developed and more versatile because gamma rays of different energies have the power of penetrating wide ranges of material and can be chosen depending on the test section used (Park and Chung, 2007). Therefore, gamma ray approaches have played a major role and have become the tools of choice in the measurement technology for gas–liquid two-phase system and gas–liquid–solid three phase system (Al-Dahhan et al., 2007; Shollenberger et al., 1997). (Wang et al., 2001) measured local porosity distribution in packed columns, the result indicated that the porosity in the column wall region is higher than that in the bulk region, due to the effect of the column wall. However, to the best of our knowledge, there are no studies reported in the literature that studied liquid holdup in two-phase upflow moving beds with the conical bottom. Attenuation of the gamma radiation is mostly due to the presence of the liquid and solid compared to gas in the flow. Thus, information on the two-phase (liquid – gas) distribution can be obtained for flow over fixed bed of catalyst, as the attenuation due to catalyst will be fixed and the variation of attenuation is due to the flowing liquid. Hence, measurement technique using gamma ray densitometry leads to measure the line averaged liquid holdup (Al-Dahhan et al., 2007; Wild et al., 1991). Accordingly, in this work a method has been developed using the gamma-ray densitometry to measure and investigate the diameter profile of the line averaged void, catalyst porosity, solid, liquid and gas holdup

and the internal liquid holdup inside the porous catalyst. For the cold flow laboratory packed bed used in this study, the column was packed randomly with a commercial extrudate catalyst. The catalyst is a porous particle which yields to an internal and external liquid holdup in the bed. The flow conditions, ranges of air and water flow rates, were kept at such levels as to simulate the industrial operation conditions of typical lab-scale hydroprocessing units. Owing to the distinct advantages of GRD, we employed it for the first time to measure the line average liquid holdup in a co-current gas-liquid upflow moving packed bed reactor operated under flowrates matching the operating conditions. The proposed correlations to predict liquid holdup or saturation are summarized in Table 1.1.

Table 1.1 Selected Liquid holdup and saturation correlations for two-phase upflow packed bed reactor.

Author (year)	Correlation
Lamine, A. S., Colli Serrano, M. T., and Wild, G., 1992	$\beta_l = (0.6\, u_g + u_l)/(u_g + u_l)$
COLLI-SERRANO AND MIDOUX, 2000	$\beta_l = 1 - (1.28 + 1.7\, u_l^{0.508} U_g^{-0.264})^{-1}$
ANIL K. SAROHA, RITESH KHERA 2006	$H_L = 0.21 + 0.00083 Re_l - 0.0026 Re_g$

2. EXPERIMENTAL SETUP

The cold flow experimental setup is scale down version of industrial upflow moving packed bed reactor. The scaled down from an industrially operated moving bed is based on dynamic and geometrical similarities. The set-up consists of a Plexiglas packed bed column, gas – supplying rotameter, liquid cycling tank, and pump. The dimensions are 57 – inch height and 11 - inch internal diameter as shown in Figure 2.1. a photograph of the experimental setup is shown in Figure 2.2. The bottom consists of two main sections, viz., gas-liquid distributor plenum and perforated cone. The plenum contains a deflector to disperse the inlet mixture of gas and liquid phases which is located at the base of the plenum to ensure that the gas and liquid are well distributed. To minimize initial maldistribution of the gas-liquid phases in the plenum and to maintain the even distribution of the two phases into the column, a gas-liquid distributor equipped with a chimney is mounted between the plenum and the cone sections. The distributor consisted of 19 holes connected with the chimneys with 0.1-inch diameter and 1.2-inch height for liquid passing through the hole. The chimney has a side hole (pitch) at the top of the chimney with 0.03 - inch diameter for gas passing. The cone section is located in between the packed bed section and the plenum section. The space between the column wall and the perforated cone wall (conical frustum distributor) contains glass marbles. Both the glass marbles and the conical frustum in the cone section are designed to provide a uniform upflow phases distribution into the catalyst bed and to maintain a stable bed operation. The packed bed section is the test section and located at the top of the cone section. The bed section was packed randomly with a 3 mm diameter industrially used porous spherical catalyst up to 24-inch as a packed bed height, and the conical frustum was used to support the catalyst. The gas-liquid flow is concurrent

and upwards through the bed particles. Tap water and oil free compressed air were used as liquid and gas feedstock in the laboratory experiment and were taken directly from the lab supply lines. Filtered compressed process air, passed through a ball valve, pressure regulator, and monitored using Dwyer Air Flowmeter, then mixed with water first before they enter the column from the bottom. The flowmeter models are accurate within ± 2% of full–scale reading. Similarly, water was pumped into to mix with air before they entered the column with the help of a liquid feed pump and returned with the air to a recirculating tank. The main purpose of this tank is to store the bypass and recycled water and vent the air to atmosphere. The mixture of gas and liquid phases are introduced to the column via an inlet pipe connected to the column base, where their streams are merged and passed upflow through the deflector. The experiments were performed at room temperature and pressure over a wide range of superficial gas velocities, and at a fixed liquid superficial velocity. The liquid velocity is selected based on the value of the scaled down liquid velocity of an industrial hydrotreating unit. The properties of the bed material and the range of operating conditions are listed in Table 2.1. Gamma ray densitometry (GRD) has been employed at various axial heights: bottom of the bed (Z/D = 0.3), and middle of the packed bed (Z/D = 1 as discussed in the following section. The horizontal measurement step was 1-inch as shown in Figure 3.1 to cover the diameter profile along the bed diameter under normal conditions and the measurement time was about 40 s in each position. For quantitative analysis of this work, each point was measured three times, and the average value was obtained of the gamma ray intensity. The horizontal and vertical measurement distance can be measured by a ruler. The experiments were run, when the stable operation was considered to have achieved, the GRD system was traversed horizontally, and scans

were performed along several chord lines parallel to the diameter of the column at any given operating conditions as shown in Figure 3.2. Eleven specified positions were equally distributed along the diameter of the column with a space interval of 1-inch. Position (0 inch) is at the center of the column and positions (5 -inch, r/R = 0.9) and (– 5 -inch, r/R = – 0.9) are the right and left horizontal positions, respectively. Liquid holdup in the packed bed was first evaluated using GRD at the same axial levels used for the actual flow scans, i.e., at Z/D = 0.3, and 1 from the bottom of the packed bed Figure 2.1, where *D* denotes the diameter of the packing and *Z* denotes the bed height. The result for the two section (bottom and middle of the bed) have been shown and discussed. Since at the top section of the bed, the catalyst has been fluidized, and the bed is not stationary, a single source cannot be used to measure the liquid holdup when the three phases are dynamically moving, where dual sources are needed (Al-Dahhan et al., 2007).

Table 2.1 System properties and range of operating conditions.

Parameter	Value / Range	ρ (kg/m^3)
Column I.D.	11 – inch	
Column Height	57 – inch	
Packed bed Height	24 – inch	
Spherical catalyst diameter	3 mm	570
Air at 25℃		1.2
water at 25℃		1000
Liquid superficial velocity, U_l	0.017 cm/s	
Gas superficial velocity, U_g	0.6 – 7.7 cm/s	

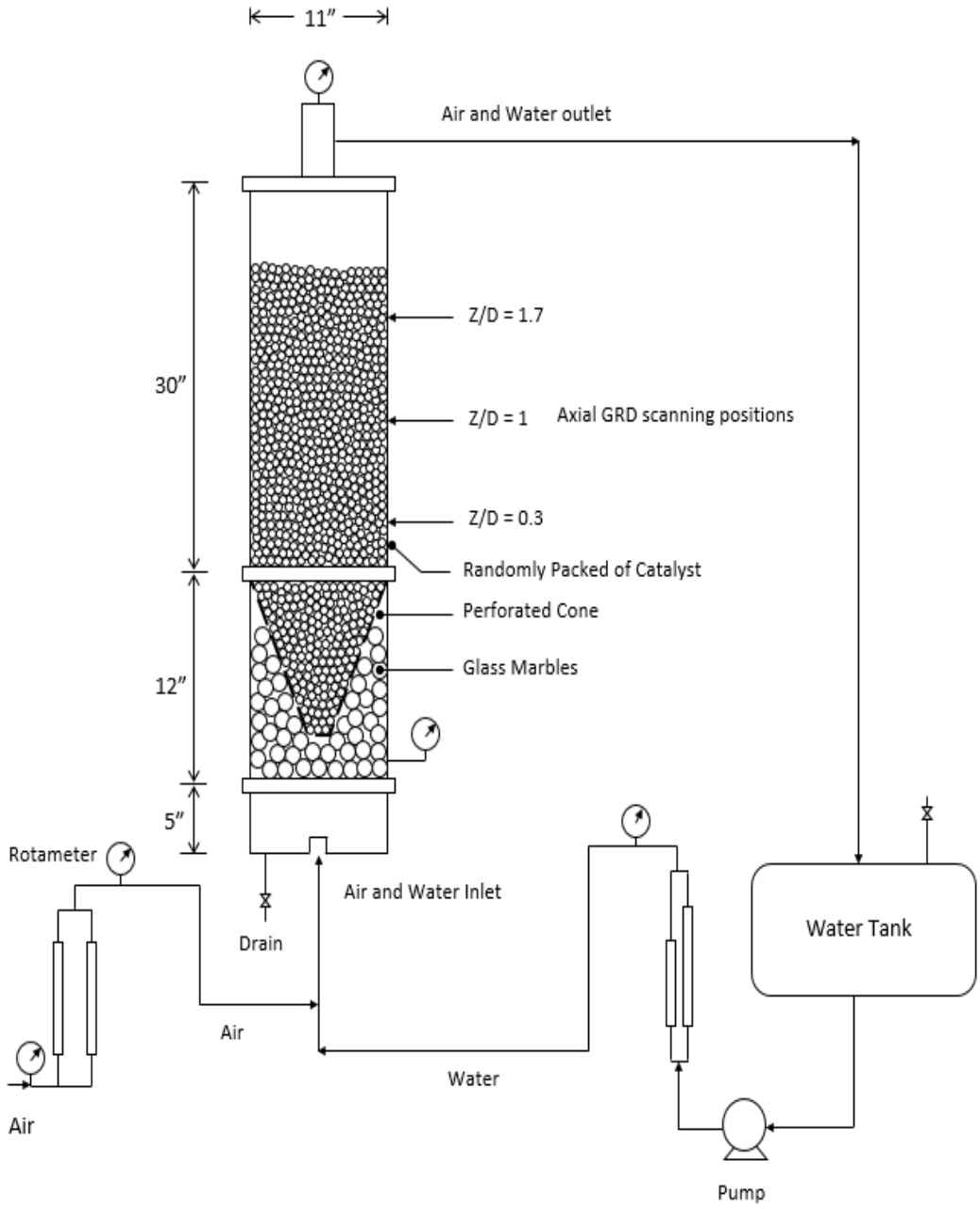

Figure 2.1 Schematic diagram of Plexiglas moving packed bed column, gas and liquid system.

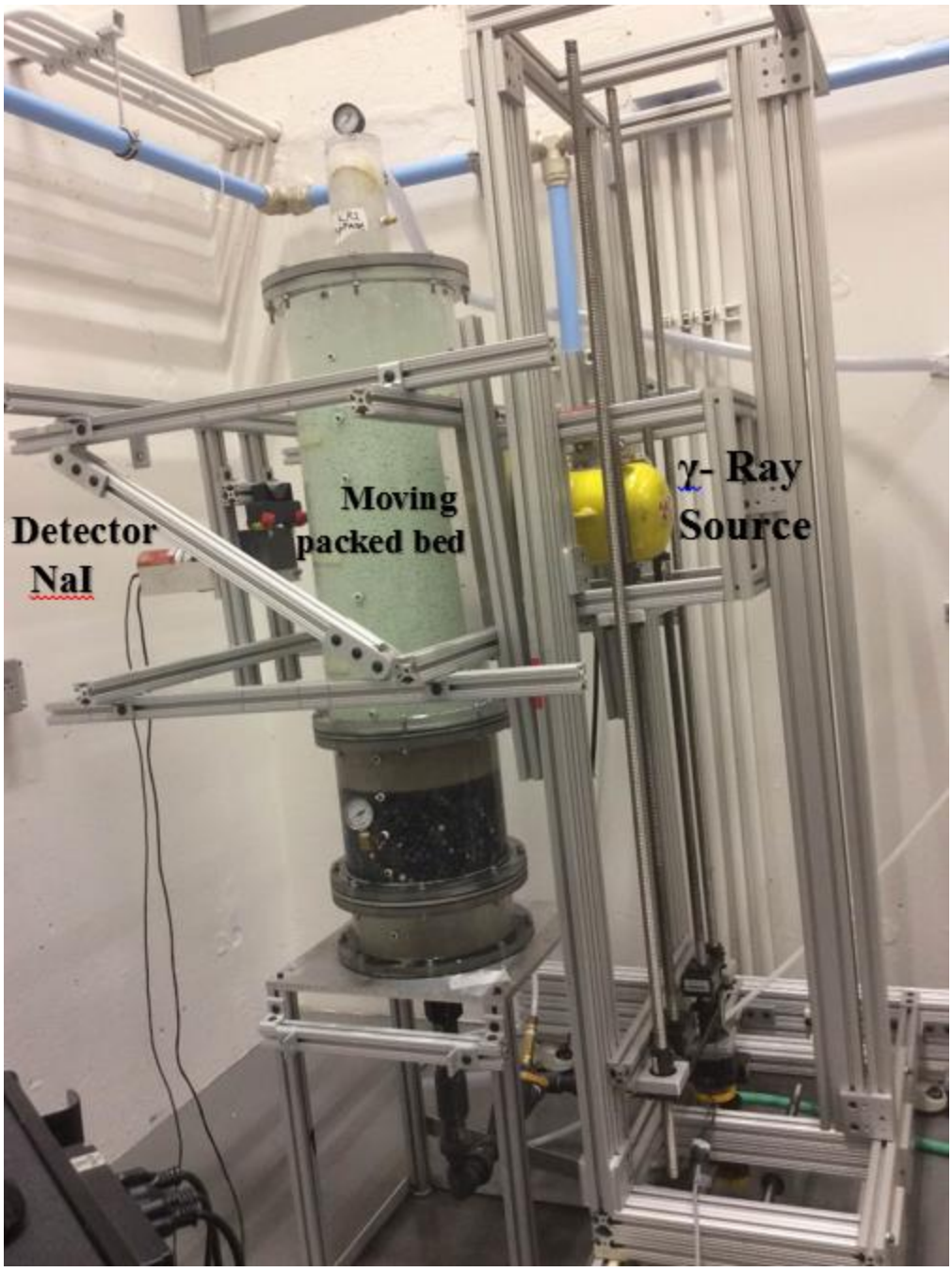

Figure 2.2 Experimental setup systems with GRD source, packed bed column, and NaI (Tl) detector.

3. GAMMA RAY TECHNIQUE

Studying the line averaged of the phase's holdup, flow regime and pattern demarcation, maldistribution identification, and online monitoring have been made possible by developing a Gamma ray densitometry (GRD) technique in our multiphase reactors engineering and applications laboratory (m-Real) at Missouri University of Science and Technology. A new methodology for the GRD technique has been developed in this study for measurement of line averaged external (in the void bed) and internal (inside the pores of the catalyst) liquid holdup. The GRD technique composes of an encapsulated 250-mCi Cs-137 gamma source with an energy of 660 keV and half-life of 30 years; thallium activated sodium iodide NaI (Tl) scintillation detector, an aluminum frame structure to support the source and detector, and data acquisition hardware and software. The ray source and the scintillation detector are both lead-shielded and placed diametrically on opposite sides of an extruded aluminum frame outside of the column. The aluminum frame is equipped with two different chain wheels to provide the vertical displacement and horizontal displacement of the source and the detector. The source-to-detector separation distance is sufficient to accommodate reactors of diameter 1.0 m. The Cs-137 source is radio-nuclide, often used in conventional gamma-ray nuclear gauge densitometers, which produces a fairly mono-energetic spectrum of gamma photons in its decay process gamma-radiation. Advantages of the long half-life of 30.17 years and appropriate emitting energy of 660 keV, Cs-137 was chosen as the common radioisotope in industrial nuclear measurements (Al-Dahhan et al., 2007). The GRD scan measurements can be made along the diameter and at any axial position of the column under study to obtain the line averaged phase distributions and holdup profiles. The packed bed column is placed at the center of

the source-to-detector separation distance. The distance from the gamma ray source to the center of the column wall is 13 cm, the total distance between the gamma ray and the detector is 57 cm as shown in Figure 3.2.

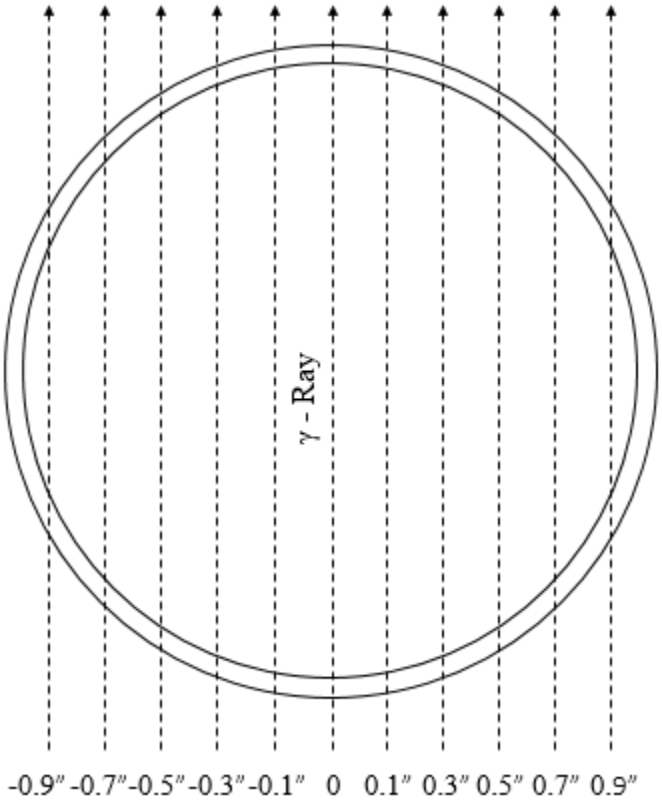

Figure 3.1 Radial scanning positions (r/R).

The photon beam of Gamma-ray coming from the radioactive encapsulated source is made such that it provides a point beam, which was custom made for the requirements of the measurement of Tracer Co Company (Pasadena, Texas). The gamma ray source is collimated by using a small diameter aperture as a radiation channel. The gamma ray then penetrate the cross-section of the experimental column section, before reaching the

collimated detector. A cubic lead collimator also collimates the detector with 4-inch length, 4-inch height and 1-inch width. Having a rectangular slot of 0.08-inch wide, 2-inch high, and 1-inch deep at a location appropriate to the detectors for sampling the radiated beam from the source. This transmitted radiation which reaches the detector contributes to the measurement. The electronic system of the NaI scintillating detector of the GRD consists of Osprey USB interface. The Osprey USB interface acts as All-in-one HVPS (higher-voltage power supply), preamplifier, and digital MCA (multi-channel analyzer), thus simplifying the electronic system. Most detectors can be represented as a capacitor into which a charge is deposited. Scintillation detector consists of a material which produces flashes of light when it absorbs radiations. Thallium-activated sodium iodide NaI (Tl) is widely used scintillators to detect the gamma rays and other ionizing radiations, the fact of its great light output among scintillators yields good efficiency and energy resolution (Khabaz and Yaghobi, 2015). The remaining gamma radiation received by the scintillation detector material generates impulse signals in response to incident radiation by a sensitive photomultiplier tube. These collected signals are amplified (The amplifier serves to shape the signal as well as further amplify it), the output signals from the amplifier discriminated for different low energy levels by multichannel analyzer (MCA), translated to a radiation intensity in the form of a measurable pulse, and transmitted to a computer as an analog signal. These signals carry information about the energy of the original incident radiation, and the ProSpect® Gamma Spectroscopy Software is used to analyze the counts received from the detector. Gamma densitometer utilizes the concept of degree of attenuation along the beam path in matter where the amount of residual radiation reaching the detector is directly related to the density of the material (i.e. a low density absorber will give less

attenuation than a high density absorber since the chances of an interaction between the radiation and the atoms of the absorber are relatively higher in latter case). Depending on a variety of flow geometries along the beam path, the amount of residual radiation that reaches the detector through the process material reflects the different flow regimes present in the column and their properties (Shaikh and Al-Dahhan, 2013). The liquid holdup measurements were made for different configuration conditions of the column to obtain the baseline data for each of the GRD scanning cases. The same measurements were later made at the same positions with liquid and flowing through the column. The average liquid holdup along each chord was then calculated based on these measurements as outlined in the next section. Using these radiation counts, which are photon counts and time series that was obtained using non-destructive inspection measurements, is analyzed to obtain meaningful results from them without any disturbance of the signal.

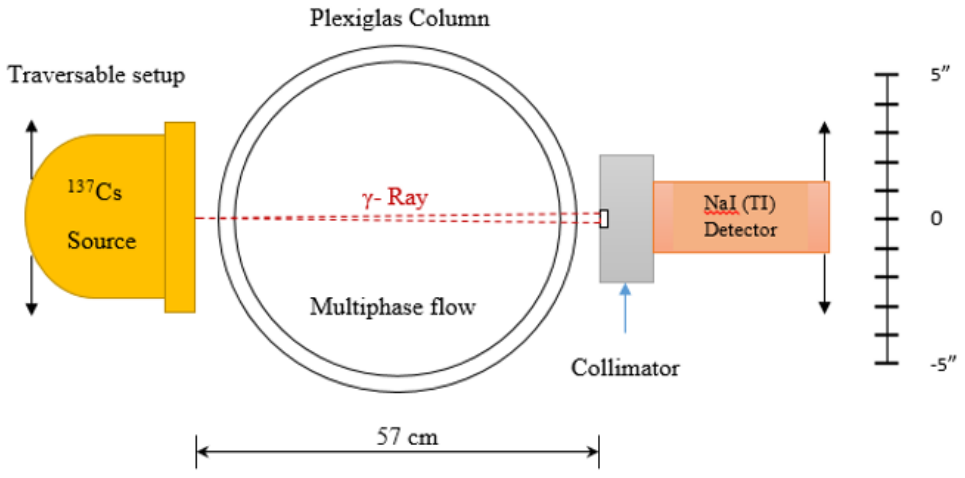

Figure 3.2 Schematic diagram of GRD showing Arrangement of Source, Plexiglas Column, Collimators and Detector.

4. PRINCIPLE OF THE HOLDUPS MEASUREMENTS AND THE NEW METHODOLOGY FOR MEASURING EXTERNAL AND INTERNAL LIQUID HOLDUPS AND CATALYST POROSITY

As mentioned earlier principle of the gamma rays densitometer technique measurement for the holdups and bed structure is based on the absorptions of gamma radiation along the beam path as it passes through the tested material (Schlieper, 2000). The attenuation of gamma ray beam depends on the radiation energy of the source, and the density and thickness of the absorbing material (Al-Dahhan et al., 2007; Jin et al., 2005). The reduction in the radiation intensity from I_0 at the source to I can be expressed by The Beer – Lambert's law according to the following equation (Chen et al., 1998):

$$I = I_0 \, e^{-\mu\rho L} \tag{4.1}$$

This equation states that the intensity of the detected radiation I is directly proportional to the intensity of the incident radiation I_0 and varies exponentially with the thickness of the absorbing medium L and its density ρ and the mass absorption coefficient μ. The attenuation ratio ($ln\frac{I_0}{I}$) called (A), and can be calculated by the natural logarithm sum of the measured attenuation I and I_0 along the gamma ray beam bath (Al-Dahhan et al., 2006) as:

$$-A = ln\frac{I}{I_0} = -\mu\rho L$$

$$A = ln\frac{I_0}{I} = \mu\rho L \tag{4.2}$$

Eq. (4.2) is the general form of the GRD beam attenuation by different material. Hence, the attenuation ratio will be different by introducing two phases inside the column.

(Schlieper, 2000) defined the incident radiation intensity (I_0), as the measured intensity of radiation reaching the detector without any absorber in between the source and the detector in the form of reference count rate (counts/s). While the detected radiation intensity (I), is different for each scan and depends on the constituting materials of the attenuating medium. Once the attenuation ratio (A) obtained for each case, the line averaged holdup of the phases can be estimated as discussed below. In our system, three phases are used where the solid phase (catalyst) is stationary while the gas and liquid are flowing co-currently upward. Hence, the attenuation ratio (A) will be the summation of the line attenuation of the individual phases. For three phases in operation of gas – liquid – solid system the attenuation ratio (A) will be:

$$A_{slg} = \ln \frac{I_0}{I_{slg}} = \mu_s \rho_s l_s + \mu_l \rho_l l_l + \mu_g \rho_g l_g \tag{4.3}$$

Where:

μ_s, μ_l,and μ_g : Mass attenuation coefficient of solid, liquid, and gas in ($\frac{cm^2}{g}$).

ρ_s ρ_l, andρ_g: Density of solid, liquid, and gas respectively in ($\frac{g}{cm^3}$).

l_s = length occupied by solid (catalyst) among the total length (cm).

l_l = length occupied by liquid (water) among the total length (cm).

l_g = length occupied by gas (air) among the total length (cm).

L = Total length that is occupied by the gas, liquid and solid along the GRD beam path including the length of air outside the column (cm) and the thickness of the column wall.

Since the path of air outside the column and the column wall are the same in each scan that we performed and these will be cancelled in the steps of manipulating the equations, the attenuation due to these medium and lengths are not included in the

following equations. Therefore, in the following equation and their manipulation only the materials inside the column of solid, gas, and liquid are included.

Where: $L = l_s + l_l + l_g$, $l_s = \varepsilon_s L$, $l_l = \varepsilon_l L$, and $l_g = \varepsilon_g L$,

ε_s,ε_l, and ε_g = the holdup for solid, liquid and gas respectively.

The attenuation ratio (A) for two phase will be:

- Gas – solid system: $A_{gs} = ln\frac{I_0}{I_{gs}} = \mu_g \rho_g l_g + \mu_s \rho_s l_s$ (4.4)
- Liquid – solid syste: $A_{ls} = ln\frac{I_0}{I_{ls}} = \mu_l \rho_l l_l + \mu_s \rho_s l_s$ (4.5)
- Gas – liquid system: $A_{gl} = ln\frac{I_0}{I_{gl}} = \mu_g \rho_g l_g + \mu_l \rho_l l_l$ (4.6)

The attenuation ratio (A) for single phase will be:

- Gas phase: $A_g = ln\frac{I_0}{I_g} = \mu_g \rho_g l_g$ (4.7)
- Solid phase: $A_s = ln\frac{I_0}{I_s} = \mu_s \rho_s l_s$ (4.8)
- Liquid phase: $A_l = ln\frac{I_0}{I_l} = \mu_l \rho_l l_l$ (4.9)

In this study, a new methodology is developed to measure the line average external void space and catalyst bed external liquid holdup in void space, and the line average internal porosity of catalyst. All GRD measurements are carried out the same axial and radial locations as mentioned in experimental setup section, with different material inside the column as described below. All GRD scans contain the attenuation value of the column wall which is constant. We removed the wall attenuation by subtracting the wall attenuation ratio (A_{wall}) from each attenuation ratio (A). The catalyst bed is fixed during our experiment except the top of the bed where the catalysts are fluidized. Therefore, the liquid

holdup, void holdup of the bed, and internal liquid holdup and porosity of the catalyst, were measured by performing the following GRD scans:

- Without column.
- An empty column for the wall attenuation.
- The column is filled only with water for the liquid attenuation.
- The column is filled with packed bed for a dry catalyst representing attenuatuoin of the gas and solid phases.
- The packed column is filled with water first, then it is drained where the scan was for a wet catalyst attenuation.
- The packed bed is filled with water representing both liquid and solid attenuation.
- Scanning the gas-liquid-solid flow under the desired operation at the same position where the holdups for all the three phases can be obtained.

GRD scanning procedures for different constituting materials and flowrate conditions were followed as reported by (Efhaima and Al-Dahhan, 2015; Rados et al., 2005; Yin et al., 2002). New methodology to measure line average liquid holdup in void space, liquid holdup inside the catalyst pores and the catalyst position for porous catalyst, has been developed and performed as follows:

1. Scannig without column (absorbing medium) I_0 (i.e. air only) In this scanning case, GRD beam passes through the atmosphere from the source to the detector without any absorbing medium in between them. The obtained attenuation I is due to air only (I_g) which represents the incident radiation (I_0) (Schlieper, 2000). The gamma ray source is placed on one side and the scintillation detector is on the other side.

2. Scannig the empty column, for wall attenuation A_c of plexiglass (air inside only, base line). In this scanning case, the GRD beam passes through the empty column, the attenuation is due to the wall of the column and the gas (air) inside it. The obtained attenuation I_c is due to wall column and air:

$$A_g = ln\frac{I_0}{I_g} = \mu_g \rho_g l_g$$

$$A_g = ln\frac{I_0}{I_0} = ln\,1 = 0$$

$$A_c = \ln\frac{I_0}{I_c} = \mu_c \rho_c l_c + \mu_g \rho_g l_g$$

$$A_c = ln\frac{I_0}{I_c} = \mu_c \rho_c l_c \qquad (4.10)$$

Where: I_c : represents the attenuation coefficient due to column wall.

The mass attenuation coefficient of the air (μ_g) is negligible compared to the Plexiglas (μ_c), less interaction of air in comparison with Plexiglas.

3. Scannig the column full of water, for liquid attenuation A_l(i.e. water inside only). In this scanning case, the same packed column filled with water only in which the GRD beam passes through the column wall and the water. The obtained attenuation is due to the wall of the column and the liquid inside it:

$$A_{lc} = ln\frac{I_0}{I_{lc}} = \mu_l \rho_l l_l + \mu_c \rho_c l_c \qquad (4.11)$$

$$A_l = A_{lc} - A_c$$

$$A_l = \mu_l \rho_l l_l \qquad (4.12)$$

By subtraction (eq.4.10) from (eq.4.11), the net attenuation of liquid (A_l) is obtained, where, the attenuation of air outside the column is neglected as illustrated in step 2.

4. Scanning the column packed with dry solid catalyst, as a $A_{ds\ g\ c}$ (i.e. dry catalyst inside only, dry solid phase). In this scanning case, the same packed column was packed with dry solid particles only in which the GRD beam passes through the column wall, the dry catalyst and the gas in voids. The obtained attenuation is due to the wall of the column, the dry solid catalyst and the gas in voids between solid catalysts, where the latter is negligible in comparison with the density of the solid along the path of the gamma ray beam from the source to the detector. By subtraction (eq.4.10) from (eq.4.13), the net attenuation of dry solid catalyst (A_{ds}) is obtained:

$$A_{ds\ g\ c} = ln\frac{I_0}{I_{ds\ g\ c}} = \mu_{ds}\rho_{ds}l_{ds} + \mu_c\rho_c l_c \tag{4.13}$$

$$A_{ds} = A_{ds\ g\ c} - A_c$$

$$A_{ds} = \mu_s\rho_s l_s \tag{4.14}$$

I_{ds} : represents the attenuation coefficient due to dry solid catalyst, and $\mu_{ds} >> \mu_g$, so $\mu_g\rho_g l_g \cong 0$

5. Scannig the column packed with wet solid catalyst, $A_{ws\ g\ c}$ (i.e. wet catalyst inside only – wet solid phase). The same packed bed, that has the dry solid catalyst particles, was filled with water for a sufficient time then the column was left to drain for a number of hours to ensure that the static liquid becomes negligible. The static holdup in the step is negligible as proper draining ensures that is no liquid outside of the catalyst pores in the measured line averaged location. Hence the only left liquid is detained inside the catalyst porous due to the capillary force (Al-Dahhan and Highfill, 1999). In this scanning case, the GRD beam passes through the column wall, the wet catalyst, and the void space of gas. The obtained attenuation is due to the wall of the column, the solid catalyst, liquid

inside the catalyst (porous), and the gas in voids between solid catalysts, where the latter is negligible:

$$A_{ws\,g\,c} = \mu_s \rho_s l_s + \mu_l \rho_l l_{int.\ \text{cat. pores}} + \mu_c \rho_c l_c \tag{4.15}$$

The obtained attenuated of the air also neglected in this case as illustrated in step2. By subtraction (eq.4.10) from (eq.4.15), the net attenuation of wet solid catalyst (A_{ws}) is obtained, Where: $\mu_g \rho_g l_g \cong 0$:

$$A_{ws} = A_{ws-g-c} - A_c$$

$$A_{ws} = \mu_s \rho_s l_s + \mu_l \rho_l l_{int.\ cat.\ pores} \tag{4.16}$$

6. Scannig the column packed with solid and liquid, $A_{l\,s\,c}$ (water – catalyst inside, liquid – solid phase).The same packed bed, contains wet solid catalyst inside, was filled with water so the voids between the particles currently filled with water. In this scanning case, the GRD beam passes through the column wall, the solid catalyst and water. The obtained attenuation is due to the wall of the column, the solid catalyst, liquid inside the catalyst (porous) and the liquid outside the catalyst in voids between solid catalysts:

$$A_{l\,s\,c} = \mu_l \rho_l l_{int.\ cat.\ pores} + \mu_l \rho_l l_{ext.\ void} + \mu_s \rho_s l_s + \mu_c \rho_c l_c \tag{4.17}$$

$$A_{l\,s} = A_{l\,s\,c} - A_c$$

$$A_{l\,s} = \mu_l \rho_l l_{int.\text{cat. pores}} + \mu_l \rho_l l_{ext.\ void} + \mu_s \rho_s l_s \tag{4.18}$$

By subtraction (eq.4.10) from (eq.4.17), the net attenuation of liquid-solid (A_{ls}) is obtained in eq. (4.18), Since: $l_{ext.\ void} = \varepsilon_\beta L$.

$$A_{l\,s} = \mu_l \rho_l l_{int.\ \text{cat. pores}} + \mu_l \rho_l\, \varepsilon_\beta L + \mu_s \rho_s l_s \tag{4.19}$$

Where (ε_β) is the line average void holdup which is completely occupied by the liquid, which is equal to the bed void.

7. Scanning the column with the desired operation of gas and liquid phases, $A_{l\,s\,g\,c}$ (air – water – catalyst, gas – liquid – solid). The packed bed contains the solid catalyst and the liquid – gas phases are introduced into the packed bed column as the desired flow operating conditions. In this scanning case, the GRD beam passes through the column wall, catalyst, liquid, and gas as a three phase liquid – solid – gas attenuation. The obtained attenuation is due to the wall of the column, the solid catalyst, liquid inside the pore, total liquid in external void (dynamic + static), and the gas phase:

$$A_{l\,s\,g\,c} = \mu_l \rho_l l_{int.\text{cat. pores}} + \mu_l \rho_l l_{ext.\,void} + \mu_s \rho_s l_s + \mu_c \rho_c l_c \qquad (4.20)$$

By subtraction (eq.4.10) from (eq.4.20), the net attenuation of liquid – solid – gas ($A_{l\,s\,g}$) is obtained:

$$A_{l\,s\,g} = A_{l\,s\,g\,c} - A_c$$

$$A_{l\,s\,g} = \mu_l \rho_l l_{int.\text{cat. pores}} + \mu_l \rho_l l_{ext.\,void} + \mu_s \rho_s l_s \qquad (4.21)$$

Where: $l_s = \varepsilon_s L$, $l_{ext.\,void} = \varepsilon_l L$, and the μ_g gas is neglected.

$$A_{l\,s\,g} = \mu_l \rho_l l_{int.\text{ cat. pores}} + \mu_l \rho_l \varepsilon_l L + \mu_s \rho_s \varepsilon_s L \qquad (4.22)$$

To measure the Bed void distribution, that represents the tortious path among the bed particles, by subtraction (eq.4.16) from (eq.4.18):

$$A_{l\,s} - A_{ws} = \mu_l \rho_l \varepsilon_\beta L = \varepsilon_\beta A_l$$

$$\varepsilon_\beta = \left(\frac{A_{l\,s} - A_{ws}}{A_l}\right) \qquad (4.23)$$

This represents the void between the bed catalyst particles that is randomly packed. The attenuation of the gas phase outside the packed bed column which is the air in this scanning work. Also, the different densities of the geometries for the absorbing material along the GRD beam bath make a difference in the attenuation of the gamma ray beam.

To measure the total liquid holdup in external void space (dynamic and static liquid holdup). Therefore, subtraction of (eq.4.16) from (eq.4.21), yields:

$$A_{l\,g\,s} - A_{ws} = \mu_l \rho_l \varepsilon_l L = \varepsilon_l A_l$$

$$\varepsilon_l = \left(\frac{A_{l\,s\,g} - A_{ws}}{A_l}\right) \tag{4.24}$$

Here due to capillary force and in the absence of surface reactions, the the catalyst pores are always intact with the liquid.

To measure the catalyst porosity fraction with respect to the total bed volume, which is equivalent to the internal liquid holdup inside the catalyst particle, subtraction of (eq.4.14) from (eq.4.16), gives:

$$A_{ws} - A_{ds} = \mu_l \rho_l l_{int.\ \text{cat. pores}} = \mu_l \rho_l \varepsilon_{int.} L$$

Where: $A_l = \mu_l \rho_l L$

$$A_{ws} - A_{ds} = \varepsilon_{int.} A_l$$

$$\varepsilon_{int.} = \left(\frac{A_{ws} - A_{ds}}{A_l}\right) \tag{4.25}$$

$\varepsilon_{int.}$: It is equivalent to the catalyst porosity fraction with respect to the bed volume.

The line average gas holdup can be measured by $\varepsilon_\beta - \varepsilon_l = \varepsilon_g$. Therefore,

$$A_{l\,s} - A_{l\,s\,g} = \mu_l \rho_l l_{int.\ \text{cat. pores}} - \mu_l \rho_l l_{ext.\ void}$$

$$A_{l\,s} - A_{l\,s\,g} = \mu_l \rho_l (\varepsilon_\beta L - \varepsilon_l L)$$

Where $\varepsilon_g = \varepsilon_\beta - \varepsilon_l$

The attenuation will be different between the phases as the densities are different, which is depending on the interaction of the gamma ray with the absorbing material. Higher density material will result in a higher attenuation of the gamma ray beam than the lower density material.

By subtraction the attenuation of the liquid-solid phase from the attenuation of the three phases, we can get the gas holdup:

$$A_{l\,s} - A_{l\,s\,g} = \mu_l \rho_l L\,(\varepsilon_g)$$

$$\varepsilon_g = \left(\frac{A_{l\,s} - A_{l\,g\,s}}{A_l}\right) \tag{4.26}$$

Then the sold holdup:

$$\varepsilon_s = 1 - \varepsilon_l - \varepsilon_g \tag{4.27}$$

5. RESULT AND DISCUSSION

5.1 DIAMETER PROFILE OF FIXED PARAMETERS OF THE BED

In this section, the line average diameter distribution of the parameters like external voidage, solids holdup, and the internal liquid holdup inside the catalyst which is equivalent to the internal porosity fraction with respect to bed volume. All these parameters are fixed for a packed bed unit irrespective of the operating conditions. The methods used to obtain these parameters have been discussed in the section of the principle of measurements.

5.1.1 Diameter Profile Of External Catalyst Bed Void Space (ε_β). Figure 5.1 shows the line average radial profiles of external bed void at the axial locations of Z/D = 0.3 and 1. The bed was randomly packed which is visible from Figure 5.1 where the void fraction varies along the measured diameter of the column. The diameter profile of void was seen to vary at the middle location less than that at the bottom of the bed. The percentage deviation calculated based on the following equation:

$$percentage\ deviation = \frac{\max(radial) - \min(radial)}{\max(radial)}$$

The percentage change with respect to maximum and minimum value is around 15 percent at the middle axial location and 13 percent at the bottom axial location. There is variation along the radial direction for both the axial location and it can still significantly affect flow distribution and efficiency of the catalyst bed (Wang et al., 2001). The randomly way in packing the catalyst inside the column can affect the diameter profile of bed voids. Higher voidage structure can create less resistance flow path and hence flow channeling and bypassing and hence flow maldistribution (Du et al., 2016).

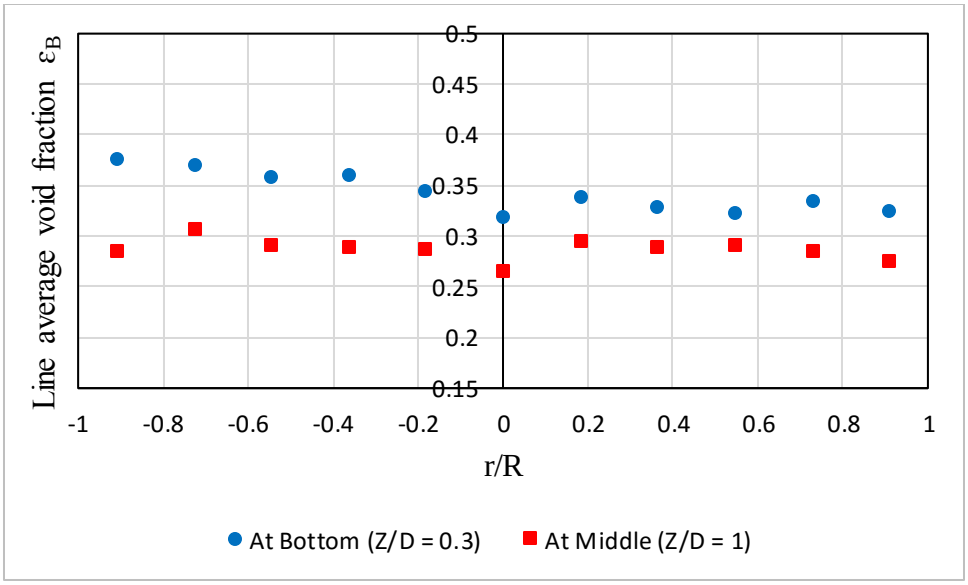

Figure 5.1 Diameter profile of external catalyst bed void space (εβ).

5.1.2 Diameter Profile Of Line Average Internal Liquid Holdup ($\varepsilon_{int.}$).

Figure 5.2 shows the line average internal liquid holdup which is equivalent to the internal porosity fraction of the catalyst particle with respect to the bed volume. This is the first time that these types of information are obtained for the porous catalyst of packed bed using Gamma-Ray Densitometry. The results indicate that the line average internal liquid holdup is not exactly same or uniform along the radial direction. The percentage deviation (section 5.1.1) along the radial direction is around 18 percent at the middle location and around 8 percent at the bottom. This can also be attributed to the fact of random distribution of packed bed, where the catalyst particles can form a small gab inside the bed. These gaps can be filled by the liquid or the gas separately, or the two phases together during the flow operation conditions. This parameter is independent of the flow rate for fixed bed reactor, and moreover, it is a function of catalyst property and bed distribution.

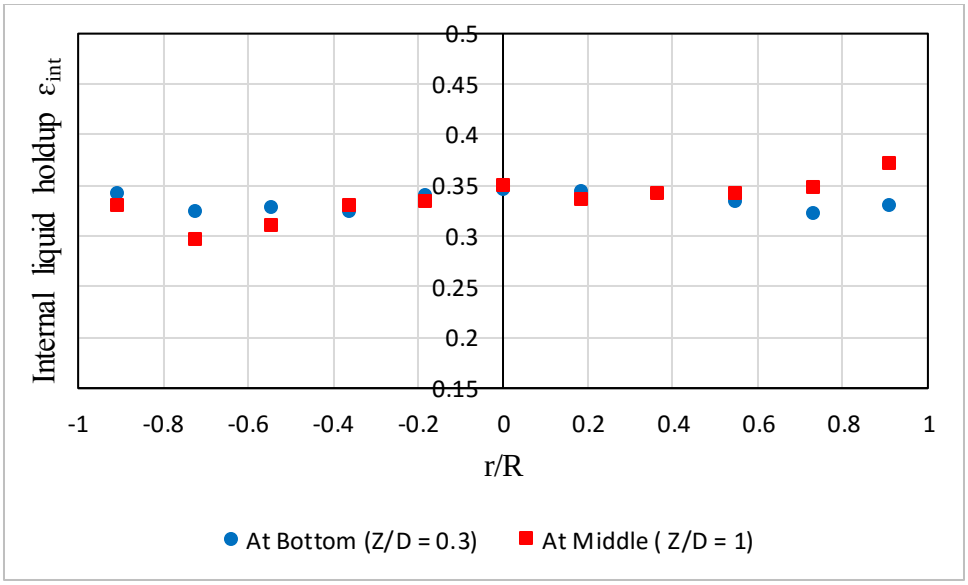

Figure 5.2 Radial distribution of line average porosity of the catalyst.

5.1.3 Radial Distribution Of Solid Holdup (ε_s). Figure 5.3 shows the line average radial profile of solid holdup and its shows random distribution and it is due to random packing and the average values is around 0.348. The percentage deviation (section 5.1.1) along radial direction at middle is around 8 percent and at bottom is around 17 percent. There is a study done to measure line average solid holdup for packed bed using GRD on random bed structure (30 cm ID column) on a similar kind of catalyst geometry (3.2 mm alumina particle) (Chen et al., 2001). Their finding shows line average solid holdup at around 0.66 but this catalyst is non-porous in nature. In our study, the catalyst is porous particles that has an internal voidage and solid matrix, therefore, the resulting summation of solid holdup (≅0.348) and line average internal porosity (≅ 0.27) gives approximate 0.61. This summation is done to ensure geometrical similarity between non-porous and porous catalyst; this also validated the method of determination of solid holdup.

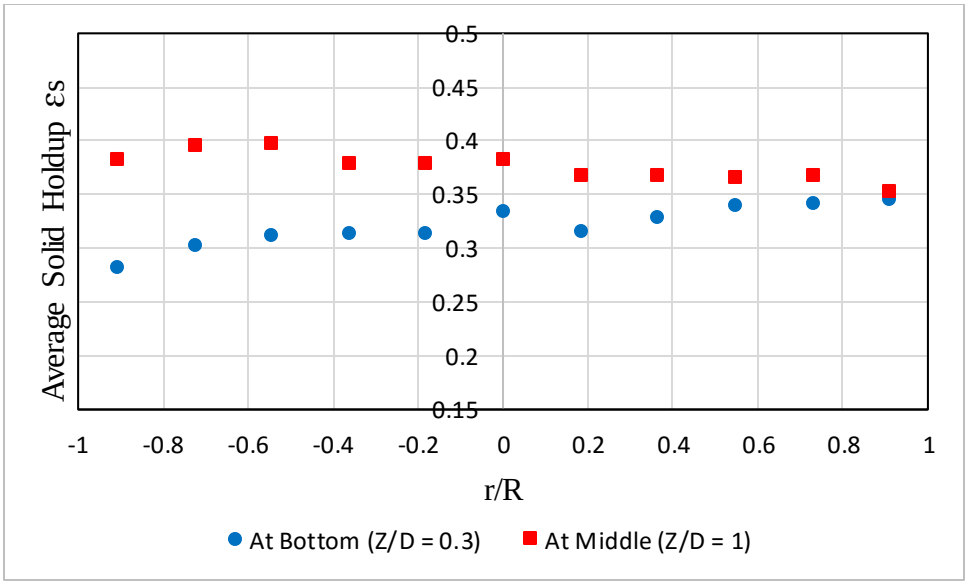

Figure 5.3 Radial distribution of Solid holdup (εs).

5.2 THE LINE AVERAGE TOTAL EXTERNAL LIQUID HOLDUPS

In this study and methodology, the measured external liquid holdup is the sum of the static liquid holdup and dynamic liquid holdup. The static liquid holdup is trapped liquid molecule in the catalyst bed, and dynamic liquid holdup is flowing liquid along the external void space of catalyst bed. For this study, the baseline operating condition is the scaled down flowrate with respect to the industrial flow rate. The gas flow rate is varied keeping the baseline liquid flow rate fixed to see the effect of the gas flow rate on external liquid holdup. The method used to obtain this parameter is showed in section 4 (principle of measurements).

5.2.1 Measurement Of Line Average External Liquid Holdup. The line average measurements of the external liquid holdup have been done on varying flow rate at fixed locations using gamma ray densitometry and scintillation detector.

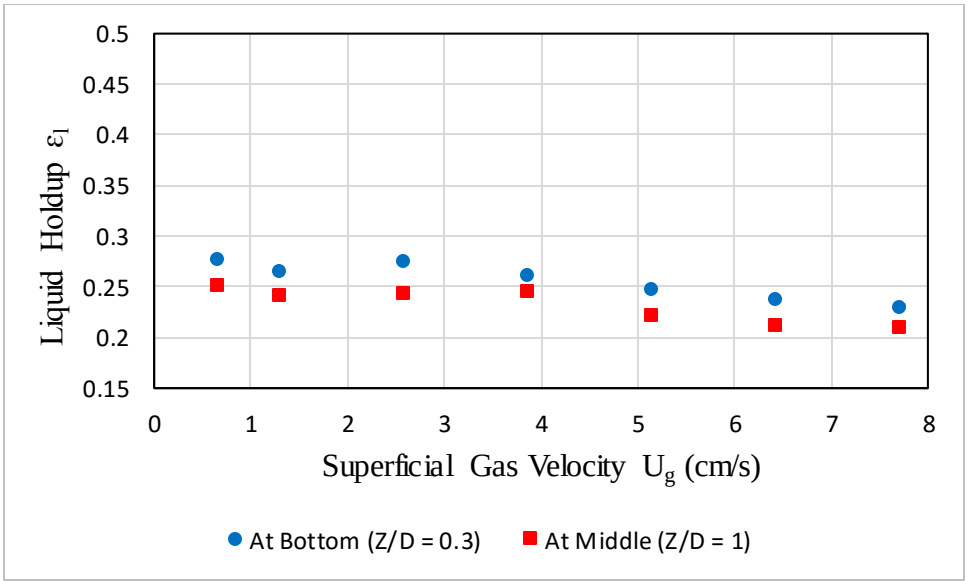

Figure 5.4 Liquid Holdup (ε_l) at the Center (r/R = 0) for the packed bed.

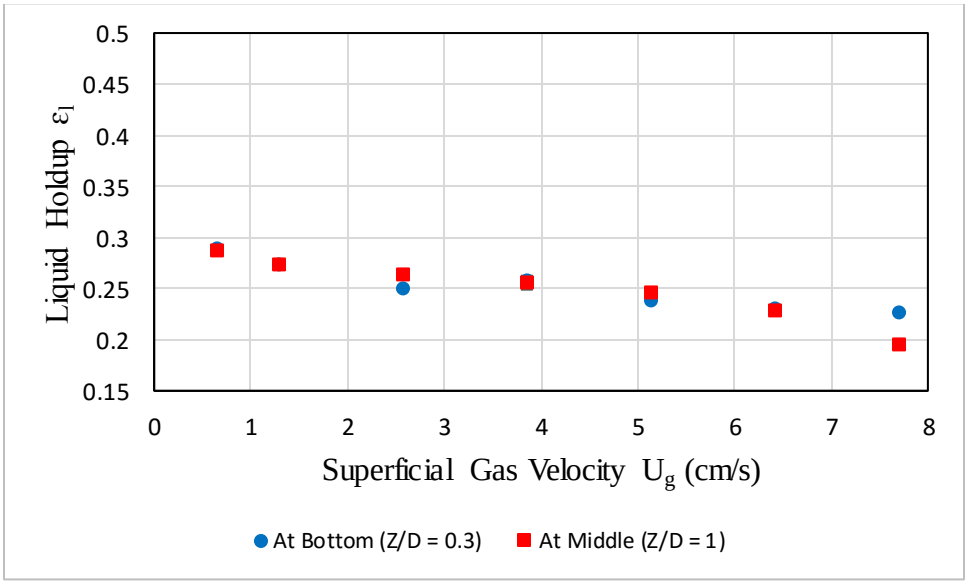

Figure 5.5 Liquid Holdup (ε_l) at (r/R = 0.5) right side of the packed bed.

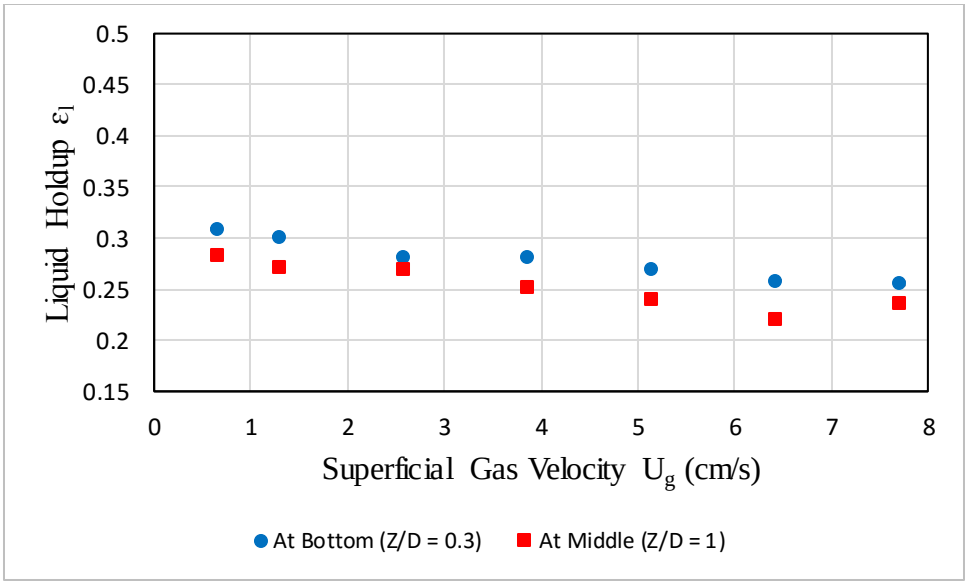

Figure 5.6 Liquid Holdup (ε_l) at (r/R = - 0.5) Left side of the packed bed.

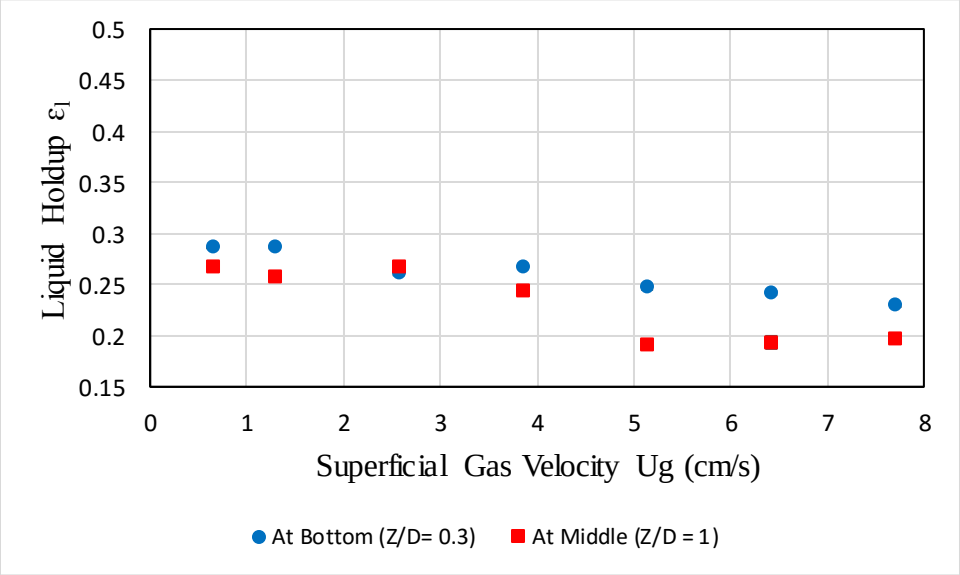

Figure 5.7 Liquid Holdup (ε_l) at (r/R = 0.9) right side of the packed bed.

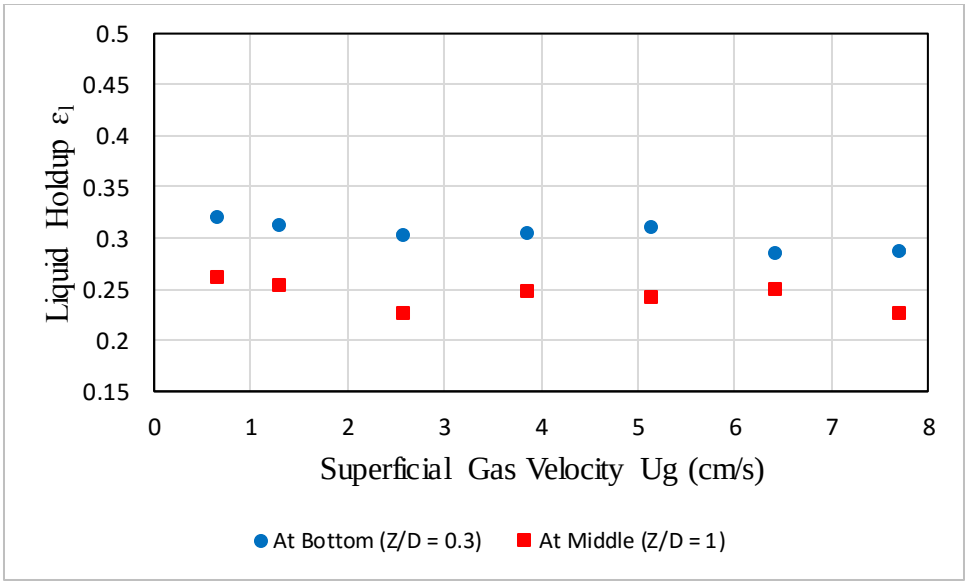

Figure 5.8 Liquid Holdup (ε_l) at (r/R = - 0.9) Left side of the packed bed.

Figure 5.4 to Figure 5.8 shows that the external line average liquid holdup (ε_l) at r/R = (0, + 0.5, - 0.5, + 0.9, - 0.9) and bottom and middle axial locations (Z/D = 0.3 and 1). It is observed that the liquid holdup is gradually decreasing, and decreasing trend is sharp after 4cm/sec for all the position except at wall (r/R = ± 0.9), it is usually seen in this reactor the transition occurs from bubble to pulse flow occurs at this flow rate "Hydrodynamic of A Co-Current Gas Liquid Upflow in A Moving Packed Bed Reactor with Porous Catalysts". Increasing gas flow rate results in a transition in the flow regime from bubbly to pulse flow for gas phase. At Bubbly flow regime the reduction of liquid holdup is not sharp, as this regime is characterized by a low interaction between bubbles themselves, bubbles and packing, and also a little effect of bed porosity and geometry on these quantities (Molga and Westerterp, 1997b). As gas velocity is increased, the fluid turbulence and the bubble number will increase, and the interference between bubbles and the coalescence/re-split will occur, which reduces bubble size and increases the gas holdup in

the reactor and then reduces the liquid holdup sharply with increasing gas flow rate. All the location irrespective of the axial and radial location showed decreasing trend of the liquid holdup with increasing gas flow rate, and the same trend is observed by (Colli-Serrano and Midoux, 2000; Kumar et al., 2012; Thanos et al., 1996). The decreasing trend calculated with respect to maximum holdup ((max-min)/max)) for all location is varying between 15 percent and 33 percent. At wall (r/R= ± 0.9) the transition of the regime is not clear due to the significant wall effect. The trend at both sides of column wall, as seen in Figure 5.7 and Figure 5.8 show quite different behavior, and this can be directly linked to the effect conical bottom and the plenums. In all the cases the liquid holdup is higher for the bottom part and this is due to external bed voidage/ external porosity of the bed structure as seen in Figure 5.1 where the void space is higher for the bottom location compared to the middle section.

5.2.2 Diameter Profile Of External Liquid Holdup. The radial profiles of the liquid holdup for the bottom part (Z/D = 0.3) of the packed bed at superficial gas velocities (U_g = 0.6, 1.2, 3.8 and 7.7 cm/s) and at constant liquid velocity (U_l = 0.017 cm/s), are shown in Figure **5.9**. The radial average is calculated for each gas velocity (Ug = 0.6, 1.2, 3.8 and 7.7 cm/s) and it is as follows ($\boldsymbol{\varepsilon_l}$ = 0.3, 0.29, 0.27 and 0.24). It seen to be decreasing as expected. The percentage deviation (section 5.1.1) for increasing flow rate is approximate as follows (11%,12%,13%,20%). At lower flow rate the radial distribution of external liquid holdup is quite uniform but on increasing the gas velocity the distribution shifts more toward one side. Where, the gas phase flows in one side and the liquid phase on the other side. This phenomenon can be due to the effect of cone base and plenums.

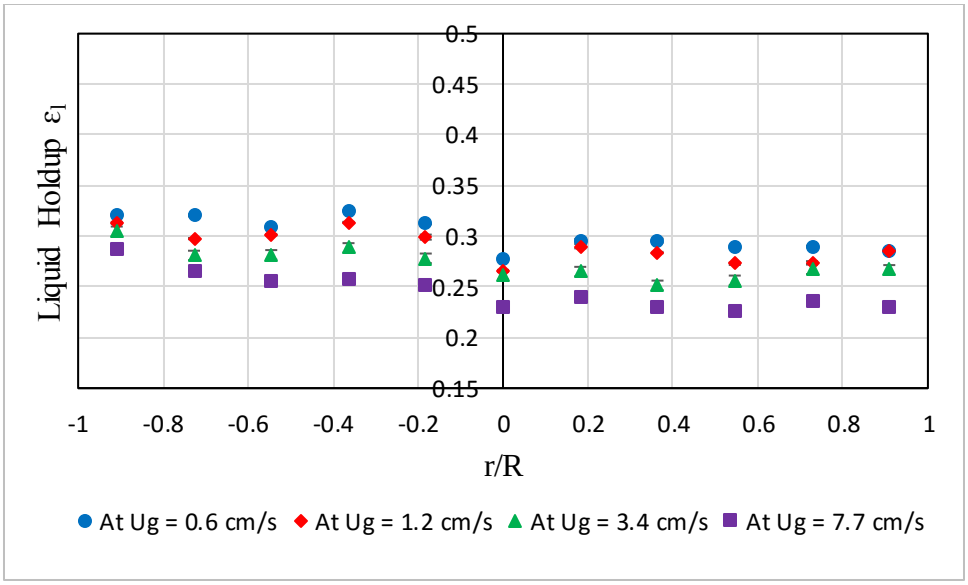

Figure 5.9 Effect of superficial gas velocity (Ug) on the liquid holdup at Bottom (Z/D = 0.3) of the packed bed at Ul = 0.017 cm/s.

Similarly, the radial profiles of the liquid holdup for the middle part (Z/D = 1) of the packed bed, are shown in Figure 5.10. It appears in Figure 5.10, a slight decrease in the average liquid holdup values obtained for superficial gas velocities (Ug = 0.6, 1.2, 3.8 and 7.7 cm/s), are (ε_l = 0.27, 0.26, 0.24 and 0.21). On comparison of the average external liquid holdup values at Z/D = 0.3 and Z/D = 1 for respective liquid velocity it is found to be quite similar expect little higher value for the bottom part. The percent deviation (5.1.1) calculated for increasing flow rate is approximate as follows (10%,11%,11%,25%) and is seen to be increasing due to more random behavior due to complex interaction of phases. At lower flow rate the radial distribution is quite uniform as also observed for the bottom part. At higher flow rate the flow distribution is better with respect to bottom part, this is due to the flow rearrangement along the axial height of bed structure.

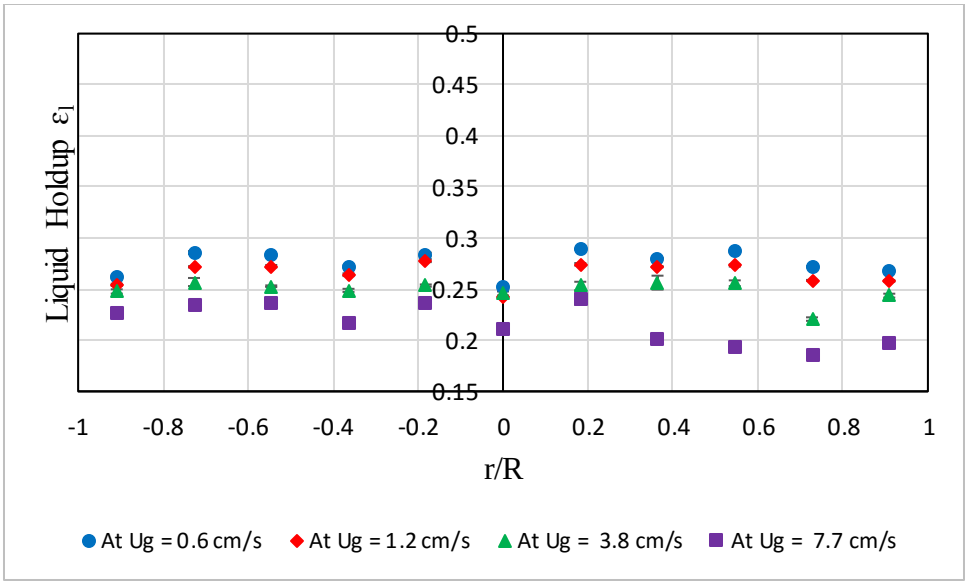

Figure 5.10 Effect of superficial gas velocity (Ug) on the liquid holdup at Middle (Z/D = 1) of the packed bed at Ul = 0.017 cm/s.

5.2.3 Comparison With Liquid Holdup Correlations. The overall liquid holdup is measured for respective superficial gas velocity by algebraically averaging values obtained at both axial and radial locations. It is compared with the correlation of overall liquid holdup for upflow packed bed with the horizontal bottom. As this is the first time studies have been conducted on upflow moving packed bed reactor with conical bottom. One study on packed bed for two phase upflow have proposed following correlation (Saroha and Khera, 2006).

$$H_L = 0.21 + 0.00083Re_l - 0.0026Re_g$$

Where: Re_l= Liquid Reynold Number and Re_g= Gas Reynolds Number.

The correlation proposed by (Saroha and Khera, 2006) gives a fair estimate of the liquid holdup and yields an average deviation of 25%.

6. REMARKS

The current study investigated the effect of varying superficial gas velocity at a constant superficial liquid velocity on the line average external liquid holdup in a co-current two phase upflow moving packed bed reactor using Gamma ray densitometry (GRD). The line average internal porosity of the catalyst particles, and line average external bed porosity have been measured also. The scanning experiments performed on an 11 - inch internal diameter upflow moving packed bed operated with an air-water system. The moving packed bed was packed randomly with 3 mm extrudate porous particles. The liquid holdup was calculated based on the new methodology developed using Beer – lambert's equation. It has been found that the liquid holdup decreased as the superficial gas velocity increased at all axial and radial locations. The rate of decrease in liquid holdup at all location except at walls is higher after 3.8 cm/sec and it is due to transition from bubbly to pulse flow regime at this superficial gas velocity "Hydrodynamic of A Co-Current Gas Liquid Upflow in A Moving Packed Bed Reactor with Porous Catalysts". The same trend observed for reported studies on upflow packed bed reactor. The external liquid holdup is higher for the bottom part than middle section of the packed bed for the same range of superficial gas velocities. At lower flow rate the liquid holdup distribution is quite uniform at both axial locations, but at higher flow rate the middle sections shows better liquid flow distributions. The result shows that the gamma ray densitometry can indicate and measure the online liquid holdup, and it's a reliable method for measuring the holdup inside packed beds with a thick wall. The comparison with available correlation on upflow packed bed showed similar trend but large absolute deviation. This necessitates further studies to develop predictable correlations for this kind of systems.

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II. IDENTIFICATION OF FLOW REGIME IN A COCURRENT GAS – LIQUID UPFLOW MOVING PACKED BED REACTOR USING GAMMA RAY DENSITOMETRY

ABSTRACT

In industries, upflow moving packed bed reactors are widely used as a leading guard reactor preceding the conventional fixed bed residual desulfurization unit (RDS). Identification of flow regimes is one of the important aspects of design, scale up, predictive model and reactor performance. Flow regime identification in this reactor was studied using gamma-ray densitometry (GRD). GRD is an important noninvasive measurement technique and flow identification can be determined by on-line monitoring. Time domain, and state space or chaotic methods are employed on photon count time series of GRD to determine flow regime. Time domain analysis includes determination of Standard Deviation, Mean, and Variance. Chaotic analyses include determination of Kolmogorov entropy (KE). All analyses are done using in-house developed programs. GRD experiments were performed on a lab scale upflow packed bed reactor built by scaling down the industrial reactor. The lab scale reactor is Plexiglas column of 11-inch I.D and 30-inch height, and it is packed randomly with 3 mm diameter catalyst till 24" height. Various axial and radial position are selected to conduct GRD scanning. The selected test location covers the bottom, middle and top of the packed bed. The measurements are conducted at superficial liquid (water) velocity 0.017 cm/s and superficial gas (air) velocity in the range of 0.6 – 7.7 cm/s. All analysis showed similar flow regime trend. When compared with flow regime map for upflow packed bed, the results indicate bubbly and pulse flow are the main regimes under this operating conditions.

1. INTRODUCTION

The recent worldwide trend in improving the quality of light fuels with the increased reserve of heavy oil have necessitated the need for designing advanced refinery processes, which are more economical, efficient, long run in combination with strict laws of environmental protection (Dorai et al., 2015; Kressmann et al., 1998). There are four main types of commercial hydroprocessing reactors that have been considered, designed and employed which are: two phase down flow fixed-bed reactors (trickle bed reactors) two phase upflow fixed bed reactors (bubble packed bed reactors), moving-bed reactors (MBR) and ebullated bed reactors (EBR). Generally, fixed bed reactor has been used for treating light petroleum feeds such as naphtha, middle distillate and heavy feedstocks < 250 ppm metal impurities. For more than 250 ppm and extra heavy petroleum feeds with higher amount of metals the moving-bed reactors and the ebullated-bed reactors have been used (Liu et al., 2009). In such a process, catalysts can lose their activity and become deactivated during the process for a variety of reasons, e.g. coke, poisoning, and sintering in which unit shutdown is unavoidable. It has been indicated, from the simulation results that gas-liquid-solid moving bed reactors could be a promising alternative in comparison with fixed bed reactors (Iliuta and Larachi, 2013b) due to a severe reduction of catalyst activity. To overcome catalyst deactivation and to attain the longest life for the catalyst, one approach is to operate the moving bed reactor, as a pretreatment system followed by the fixed bed reactor. Moving bed reactor offers a relatively large catalyst migration time, in comparison with the liquid and gas phases mean residence times, and hence, can be considered as a pseudo-two- phase gas-liquid upflow fixed bed where the catalysts are periodically removed and supplied to the reactor. Therefore, for the sake of hydrodynamics

study, moving bed reactor is investigated as two-phase uflow packed bed reactor. The only difference in this case is the configuration and the design of the bottom part of the bed which takes a cone shape to facilitate the removal of the catalyst and to distribute the upflow of gas and liquid phases. While there are a number of studies reported in the literature on the packed bed upflow reactors, where the bottom is a flat perforated or chimneys mounted plate for the catalyst support, and for distributing the upflow of gas and liquid phases. There is no study in the open literature related to the packed bed upflow configuration of moving bed with a cone design bottom. It is generally recognized, that various flow regimes exist in co-current upflow packed bed reactors, such as bubble flow, spray flow, and pulse flow regime (Shah, 1979). The reactor performance, volume productivity, mixing characteristics, flow distribution and the mass and heat transfer processes within a reactor depend strongly on the prevailing flow regimes (Nedeltchev, 2015; Thome, 2004). Hence, it is important to know how to identify the flow regime (Nedeltchev et al., 2003). In packed bed reactors, the flow regime depends on the bed parameters, liquid physical properties, particle size, and the fluid flow rates (Colli-Serrano and Midoux, 2000). Identification of these flow regimes in a packed bed is necessary in order to ensure the desired flow regime for the selected gas-liquid reaction. The results are often displayed in the form of a flow regime map that identifies various flow regimes with respect to flow and physical properties parameters such as for example the proposed flow regime map of Fukushima and Kusaka (1977). Flow regime maps can be obtained using different experimental methodologies, such as by monitoring either a sharp increase in the pressure fluctuations, sudden changes in the gas – liquid mass transfer coefficients and analysis of variations in the apparent electrical conductivity of the bed, or the analysis of

flow images obtained using capacitive sensors (Moreira and Freire, 2003b). Techniques employed for identifying the flow regimes commonly measure simple pressure fluctuation signals, two-phase electric conductivity probe signals, or visual observation through a transparent wall (Revankar et al., 2007). There are limitations with using the visual observation as it requires a transparent wall and a transparent liquid while at the wall. Hence, visual observation is not always applicable especially with opaque reactor or with combination of multiphase gas – liquid – solid that is also effectively opaque. Also, at higher flow rates the visual observation might not be very reliable as well, because only the vicinity of the wall can be observed. Pressure drop technique can give global information at the wall for the flow regime measurement and it may not reflect what happens inside the bed. For packed bed reactors, the limitation to use this measurement technique is a care should be taken in order no solid particles are in direct contact with the membrane of the pressure sensor (Boyer et al., 2002) besides the measurement is at the wall. (Raghavendra Rao et al., 2011) classified the flow regimes in the upflow packed bed into two main classifications: first, single phase pore flow and two phase pore flow, and second, bubble flow, pulse flow, spray flow, and different names for the intermediate regimes. Bubble flow, pulse flow and spray flow also were reported by other researchers (Lamine et al., 1992a; Molga and Westerterp, 1997a; Moreira and Freire, 2003b; Varma et al., 1997). The single phase pore flow happens at low gas flow rates when the interstices pores between the packing particles are predominantly covered by either of the phases separately, while the two phase pore happens at high gas flow rates when the interstices pores between the packing particles are filled by both phases. These kinds of pore flow regimes are observed when the sizes of the column packing particles were small or equal

to 2 mm in diameter , while bubble flow, pulse flow and spray flow have been observed for packing particles greater than 2 mm (Raghavendra Rao et al., 2011). Bubble flow exists at low gas flowrate and it's characterized by dispersed gas flows as bubbles in the continuous liquid phase. Pulse flow occurs at higher gas flowrate and it is characterized by liquid rich waves or "pulses" followed by a gas rich portion traverse upwards through the column length at almost regular time intervals. These pulses result in local fluctuations in liquid holdup, pressure drop, and heat and mass transfer rates. Hence, pulse flow regime enhances overall heat and mass transport while reducing axial dispersion, making it a potentially attractive mode of operation (Wilhite et al., 2005). When the reactor operates in pulse flow regime, the rate of the reaction will enhance and increase in value up to 30% while holding the other parameter in the reactor at constant (Wilhite et al., 2005). Spray flow takes place at highest gas flowrates and it is characterized by continuous gas flow with dispersed liquid flowing as film over the particles and partly as droplets in the continuous gas phase. Therefore, the proper understanding and identification of these regimes are crucial for designing and operating any reactor. Only few investigations of flow regime identification for co-current gas liquid upflow regular packed beds, with a plate at the bottom to support the catalyst and to distribute the phases, were published compared to co-current downflow packed beds (trickle bed reactors). Investigation of flow regime identification for co-current gas liquid upflow packed bed are: (Raghavendra Rao et al., 2011) identified the flow regime in two phase upflow packed bed reactor, Bubble flow, pulse flow and spray flow were identified by visual observation in their study. (Moreira and Freire, 2003b) identified the flow regime by visual observation and they described three flow regimes: bubble flow, transition I flow, pulse flow and spray flow.

(Murugesan and Sivakumar, 2002) observed the bubble flow regime as uniform bubbles flow in liquid, dispersed bubble flow regime as uniform and tiny bubbles flow in liquid and pulse flow regime. (Colli-Serrano and Midoux, 2000) reported bubble flow, transition flow and pulse flow regime. Their experiments were carried out at low gas flowrates, so they didn't detect the spray flow regime. The transition from bubble flow to pulse flow regime was identified at (0.05 kg/m^2 s) air flow rates and water flow rate range 1 – 10 kg/m^2 s. (Iliuta and Thyrion, 1997) identified two flow regimes for upflow packed bed: bubble flow and pulse flow regimes for two systems air – water and air – Carboxymethyl-cellulose (CMC). They mentioned that the transition from bubble flow to pulse flow was unclear for upflow mode of operation. Instead, they noticed a common zone of transition ($0.092 < G < 0.12$ kg / m^2 s). (Varma et al., 1997) identified bubble flow, pulse flow and spray flow regimes for bed of a ceramic spheres they used. They presented a criteria for transition from a regime to another.(Molga and Westerterp, 1997a) observed by visual observation the bubble flow regime and churn and pulsation flow regime at highest gas velocity. (Lamine et al., 1992b) identified for glass spheres of 4 and 6 mm: bubble flow, pulse flow and spray flow regimes. For smaller packing of 1 and 2 mm, they observed a regime which is termed a separated flow in the pores or single phase pore flow. They mentioned the transition between from bubble to pulse flow occurred for gas flow rates range 0.1 – 0.15 kg/m^2 s. Most of the reported studies above used the visual observation to identify the flow regimes. However, radiation based techniques have been used in a wide range of applications as measuring and monitoring techniques, since they are generally considered non-intrusive, independent of the environment, temperature and pressure of the system, non-contact with the media and continuous in their measurements (Khorsandi and

Feghhi, 2011). Radiation methods include the use of fast neutron scattering and attenuation techniques as well as the use of gamma and X-ray attenuation, and fast neutron scattering. Compared with other radiation techniques, the gamma ray technique is more versatile due to its penetration power different energies can be chosen and used safely depending on the test section (Park and Chung, 2007). Among many gamma ray based techniques, nuclear gauge densitometry which is called here gamma ray densitometry is one of the most non-invasive techniques used on industrial scale units as a diagnostic tool and on-line level monitoring device (Khorsandi and Feghhi, 2011; Shollenberger et al., 1997; Shaikh and Al-Dahhan, 2013). (Park and Chung, 2007) used single beam gamma ray densitometry to study the average void fraction in a 10.9 mm diameter stainless steel pipe under critical flow conditions. (Wang et al., 2001) measured local porosity distribution in packed columns, using single beam and scintillation detector. (Shaikh and Al-Dahhan, 2013) used single beam gamma ray to identify flow regime in bubble column. As the actual flow regimes are time varying combinations of different flow regimes; Therefore, flow regimes can also be identified by the use of only one source/detector module (Tjugum et al., 2002). The attenuation of the gamma ray passing through the bed of catalyst with upflow of gas and liquid depends on the nature of the flow conditions and on the holdups of the liquid / gas phases along the line of the gamma ray. The dynamic variation of the flow patterns and phases holdups depends on the type of flow regime. The reactor is operating at gas superficial velocity (0.6 – 7.7 cm/s) and at constant liquid superficial velocity (0.017 cm/s). Therefore, the photon counts of the measured time series fluctuate depending on the type of the flow regime and pattern that the gamma ray beam is passing through. Accordingly, the focus of this work is to implement the gamma ray densitometry (GRD) technique to

identify the flow regime in an upflow moving packed bed reactor by processing chaotically and statistically the measured time series of the photon counts. The gamma ray source and the scintillation detector are aligned externally of the packed column. Thus, the measurements were taken without disturbing the flow operation conditions inside the packed column.

2. EXPERIMENTAL WORK

2.1 EXPERIMENTAL SET-UP

A packed bed with the bottom design that represents a selected design of industrial moving bed has been developed to conduct the investigation on a flow regime and its identification of these types of reactors using gamma ray densitometry. Hence, our cold flow scale down experimental set–up used in this work consists of a Plexiglas packed bed column, a representative bottom configure of a cone type of a moving bed, gas – supplying and rotameter, liquid cycling tank and rotameter, and a pump as schematically shown in Figure 2.1. The bed and its bottom were scaled down from and industrially operated moving bed based on dynamic matching pressure drop and geometrical similarities. The laboratory packed bed of 145 cm height and 27.94 cm internal diameter consists of three main sections, which are gas – liquid distributor plenum section, cone section, and packed bed section as shown in Figure 2.1. The plenum contains a deflector to disperse the inlet mixture of gas and liquid which is located at the base of the plenum to ensure that the gas and liquid are well distributed. To minimize initial maldistribution of the gas-liquid phases into the packed bed, a gas-liquid distributor equipped with chimneys is mounted between the plenum and the cone section. The distributor consisted of 19 holes connected with chimneys with 0.25 cm diameter and 3 cm height for liquid passing through their bottom openings. Each chimney has a side hole (pitch) with 0.07 cm diameter for gas passing through the top as the gas phase forming a top gas layer at the top of the plenum. Both the deflector and the distributor plate with chimneys are distributed on an integral part of distribution section where it is followed by the cone section. The cone section is located in between the packed bed section and the plenum section. The cone section contains glass

marbles and a conical frustum distributor. Both the glass marbles and the conical frustum in the cone section are designed to provide a well distributed upflow phases and to maintain a stable bed operation. The packed bed section is the test section which consists of the packing particles (catalyst) that fill the cone and the top cylindrical column mounted at the top of the cone section. The gas-liquid flow is co-current and upwards through the bed particles. The experimental measurements were performed on the cylindrical section of the bed which was packed randomly with a 3 mm diameter spherical catalyst up to 63 cm height. These spherical catalyst particles also fill the perforated frustum cone that was used to support the catalyst as shown in Figure 2.1. Water and oil free compressed air were used as liquid and gas phases. Filtered compressed process air, passed through a ball valve, pressure regulator, and monitored using Dwyer Air Flowmeter before entering the column from the bottom. The flowmeter models are accurate within ± 2% of full–scale reading. Similarly, water was pumped into the column with the help of a liquid feed pump and returned with the air to a recirculating tank. The main purpose of this tank is to store the bypass and recycled water and vent the air to the atmosphere. The gas and liquid phases are introduced to the column via an inlet pipe connected to the column base, where their streams are merged and passed upflow through the deflector. The experiments were performed at room temperature and pressure over a wide range of gas superficial velocities and at fixed liquid superficial velocity. The properties of the bed material and the range of operating conditions are listed in Table 2.1. The scaled down liquid and gas superficial velocities are 0.017 cm/s and the range for gas (0.6 – 7.7 cm/s) respectively. These have been selected to represent the operation of the industrial moving bed which it is at incipient fluidization. Therefore, in this work, the superficial liquid velocity was kept constant at

0.017 cm/s while the superficial gas velocity was varied from 0.6 to 7.7 cm/s. Gamma ray densitometry (GRD) technique which provide line beam of gamma ray has been used to measure and identify flow regime by processing chaotically and statistically the time series of the attenuated gamma ray that is received as counts by the detector. The received photon counts through the lead collimator in front of the scintillation detector are only contribute to the measurements. The received photon counts vary in value due to the variation in the geometries inside the column. As the density of the absorbing material vary along the path of the gamma ray beam, the received photon counts vary accordingly. Higher density material will result in more attenuation for the gamma ray beam which depending on the interaction of the gamma ray beam with the atoms of the absorbing material.

Table 2.1 System properties and range of operating conditions.

Parameter	Value / Range	ρ (kg/m^3)
Reactor I.D.	27.94 cm	
Reactor Height	145 cm	
Packed bed Height	63 cm	
Spherical catalyst diameter	0.3 cm	570
Air at 25℃		1.2
water at 25℃		1000
Liquid superficial velocity, U_L	0.017 cm/s	
Gas superficial velocity, U_g	0.6 – 7.7 cm/s	

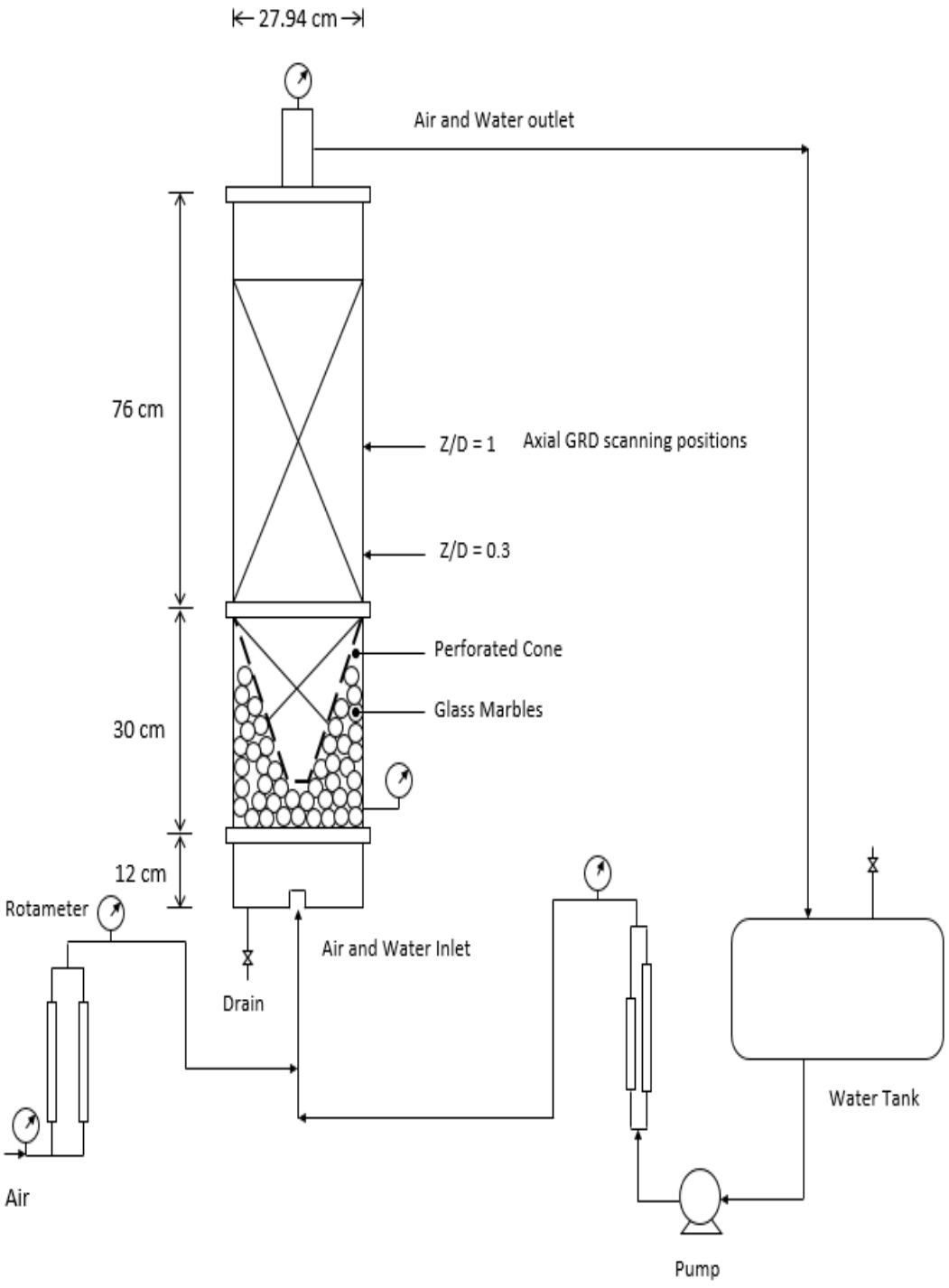


Figure 2.1 Schematic diagram of Plexiglas moving packed bed column, gas and liquid system.

2.2 THE USE OF GAMMA RAY DENSITOMETRY FOR FLOW REGIME IDENTIFICATION

In this work, identification of the flow regime has been made by implementing a Gamma ray densitometry (GRD) technique developed in our multiphase reactors engineering and applications laboratory (m-Real) at Missouri University of Science and Technology. The radiation based techniques such as gamma ray densitometry (GRD) are commonly used in a wide range of applications as measuring methods, since they are generally considered relatively simple, not expensive, and non-intrusive. It appears as a continuous measurement, non-contact and its independent performance from environment, temperature and pressure (Khorsandi and Feghhi, 2011). Among the many radiation techniques, the gamma ray technique is well developed and have played an important role in measurement technology for multiphase system which is applied in many industrial fields including petroleum industry (Khorsandi and Feghhi, 2011; Shollenberger et al., 1997; Shaikh and Al-Dahhan, 2013; Park and Chung, 2007). The principle of GRD measurement technique is based on the attenuation of the gamma ray beam depending on the density of the tested medium. The gamma ray densitometry used in this study for identification of flow regime is composed of an encapsulated 250-mCi Cs-137 gamma source with appropriate emitting energy of 660 keV, thallium activated sodium iodide NaI (Tl) scintillation detector, an aluminum frame structure to position the source and detector, and data acquisition hardware and software. The Cs-137 gamma source and the NaI (Tl) scintillation detector are both lead-shielded and placed diametrically on opposite sides of an extruded aluminum frame externally of the tested column. Thus, the GRD scan measurements can be made along the diameter and at any axial position of the column under study to identify the flow regime in this work. The horizontal and vertical

measurement distance can be measured by a ruler. The horizontal measurements and then results that is presented where made at r/R = 0, ± 0.5, and ± 0.9 as shown in Figure 2.2, and the measurement time was about 2 mins in each position. The experiments were run, when the stable operation was considered to have achieved, the GRD system was traversed horizontally and scans were performed along the chord lines mentioned above parallel to the diameter of the column at any given operating conditions. These measurements have been done at two axial heights bottom of the bed (Z/D = 0.3), and middle of the packed bed (Z/D = 1).

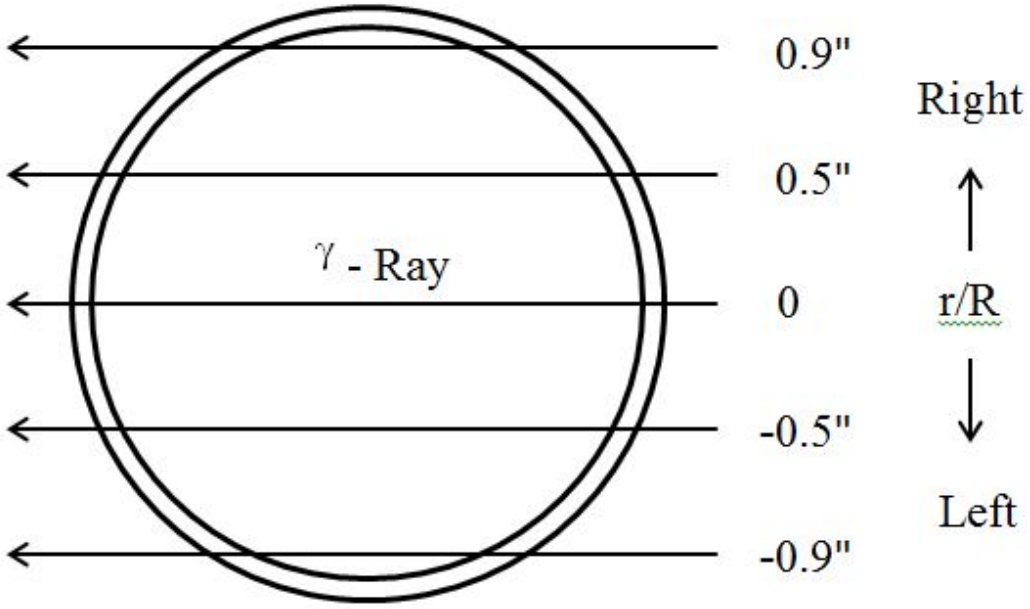


Figure 2.2 Radial scanning positions (r/R).

This transmitted radiation, that reaches the detector as time series of photon counts have been processed chaotically and statistically to identify the flow regimes and their transition since the attenuation and its dynamic fluctuations of the transmitted gamma ray vary depending on the type of flow patterns and flow regimes.

2.3 METHODS OF PROCESSING OF THE GRD SIGNALS AND DATA ANALYSIS

In the present study, the time series signal of the photon counts measured by the gamma ray densitometry in the moving packed bed at various conditions have been processed using both state space analysis (chaotic analysis) and the statistical analysis to identify the flow regimes as follows:

2.3.1 Time Series Of The Photon Counts. In this work, the time series has been taken for over 2 minutes long of data acquisition at 50 Hz that generated ≅ 3000 points for each measurement. To demonstrate the nature of dynamic fluctuations of the photon count and how they vary with the change of the flow conditions, we present here the first 100 points to show how it looks like the print of the signal. As shown in Figure 2.3 to Figure 2., the signals consist of a sequence of time series of photon count measured by NaI scintillation detector at various conditions. The GRD stet up consist of a scintillation detector (NaI) with a radiation source (Cs-137) on the opposite side. The amount of attenuated radiation reached the scintillation detector after passed the material depends on the density of that flow inside the column. The dynamic fluctuation of the attenuated radiation possess information about the flow behavior of the flow inside in the column and its properties (Nedeltchev, 2015; Shaikh and Al-Dahhan, 2013). These are a data point, we collected approximately 3000 points these 3000 points at frequency 50 Hz, and the Δt measurements = 1/frequency. The fluctuation measurements have been done by varying the flow rate conditions using gamma ray densitometry. The radiation source and the scintillation detector were aligned externally of the column. The gas superficial velocity was varied from 0.6-7.7 cm/s at a constant liquid superficial velocity.

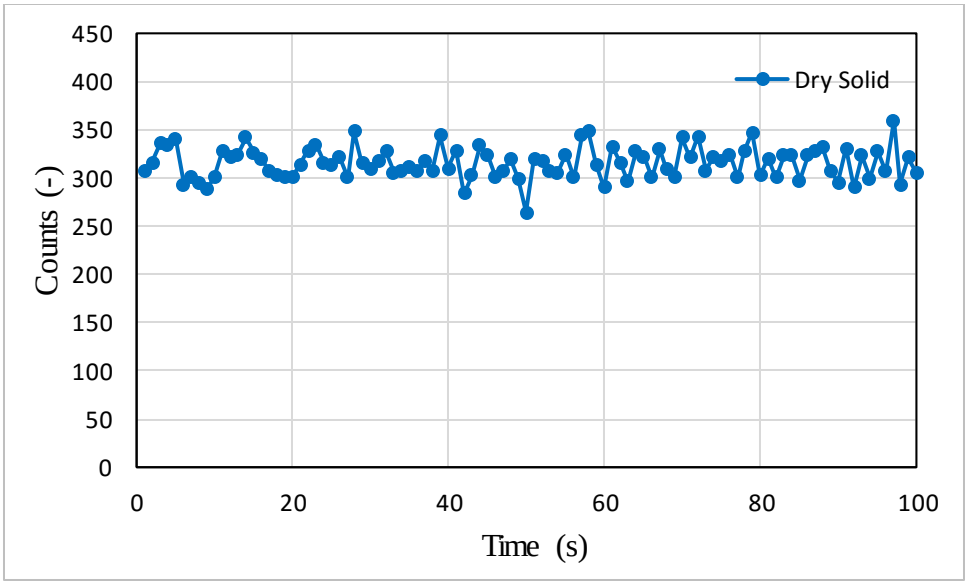
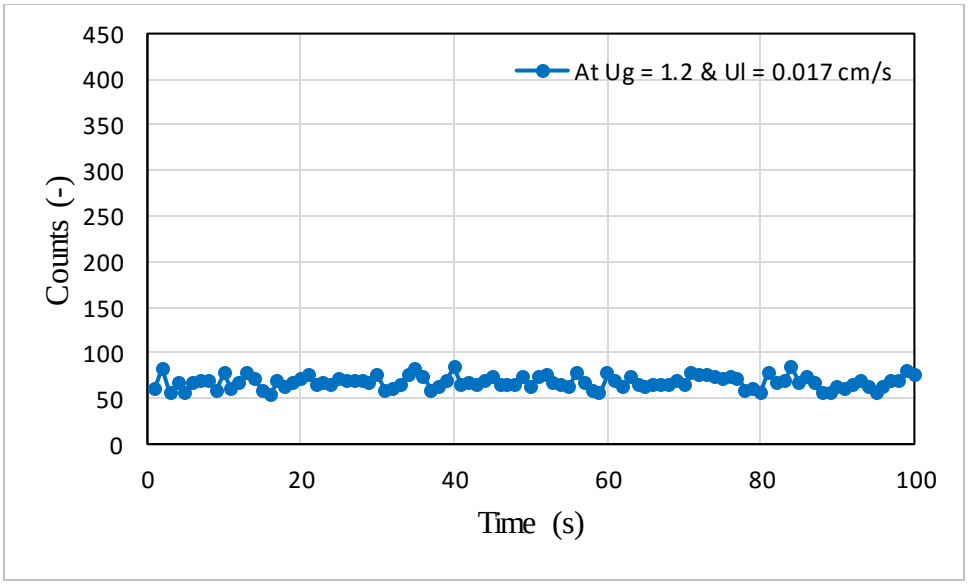
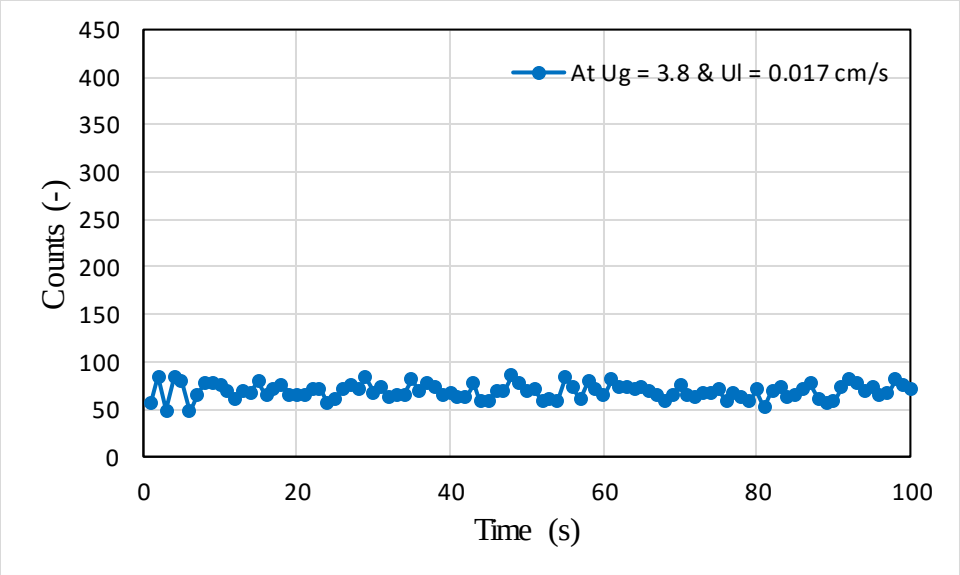


Figure 2.3 The segment of the time series of photon count fluctuations recorded in a moving packed for dry solid.

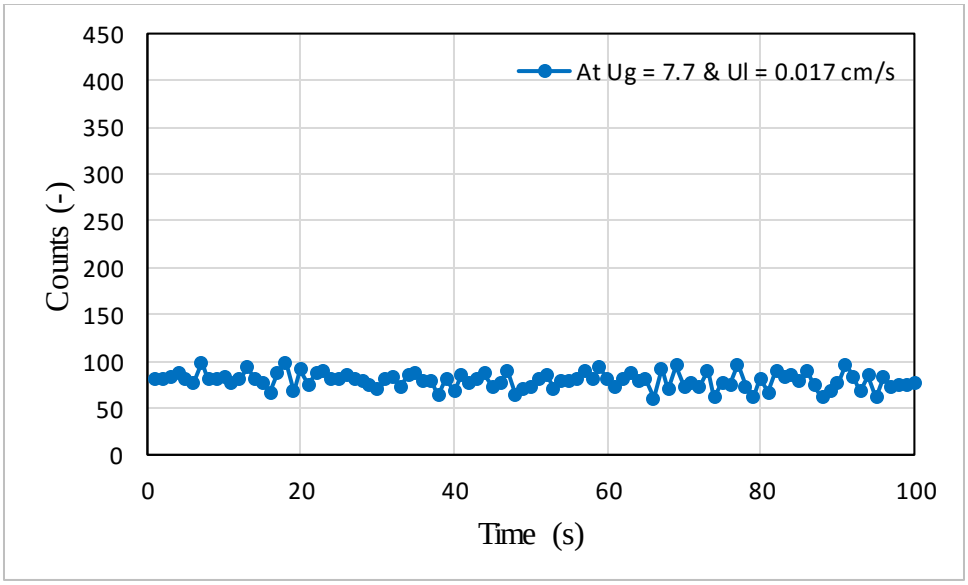


(a)

Figure 2.4 The segment of time series of photon count fluctuations recorded at Bottom (Z/D = 0.3 and r/R = 0) for the liquid superficail velocity of 0.017 cm/s and gas superficial velocity: (a) Ug = 1.2 cm/s. (b) Ug = 3.8 cm/s. (c) Ug = 7.7 cm/s.

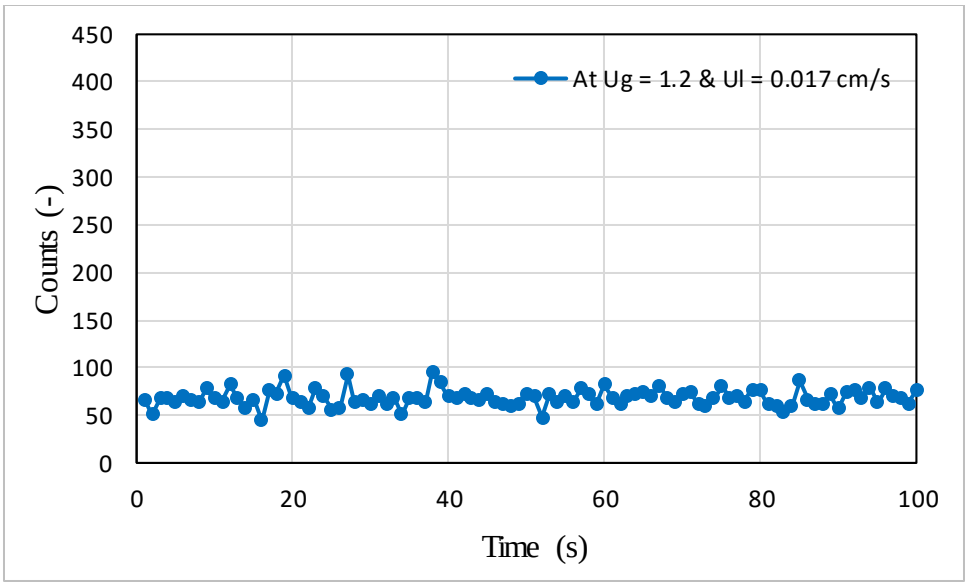


(b)

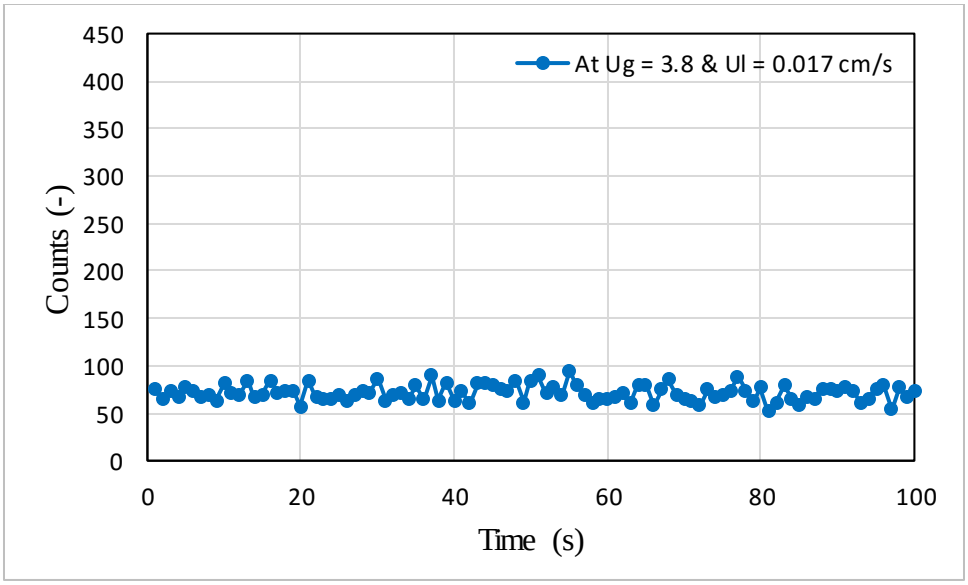


(c)

Figure 2.4 The segment of time series of photon count fluctuations recorded at Bottom (Z/D = 0.3 and r/R = 0) for the liquid superficail velocity of 0.017 cm/s and gas superficial velocity: (a) Ug = 1.2 cm/s. (b) Ug = 3.8 cm/s. (c) Ug = 7.7 cm/s (cont.).

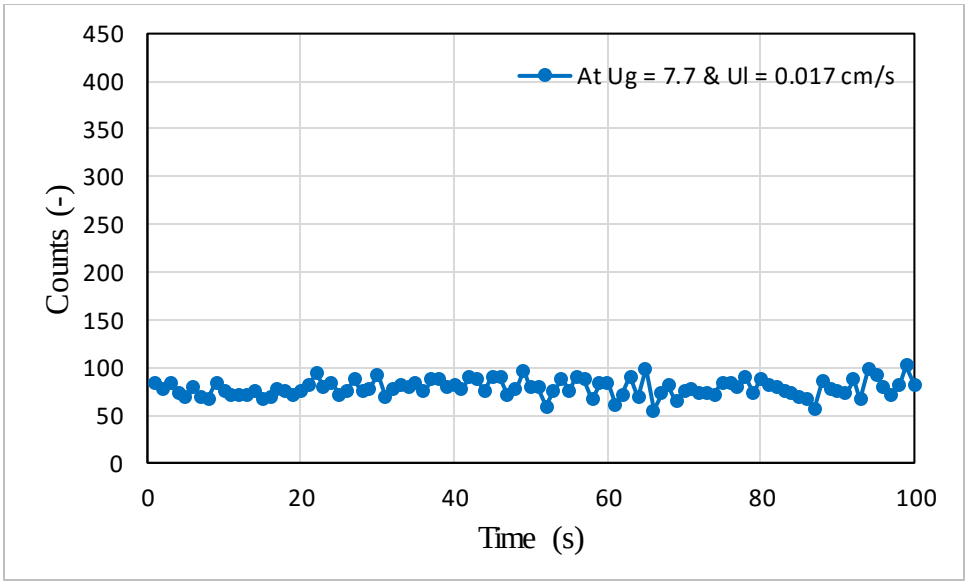


(a)



(b)

Figure 2.5 The segment of the time series of photon count fluctuations recorded at Middle (Z/D = 1 and r/R = 0) for the liquid superficail velocity of 0.017 cm/sec and gas superficial velocity: (a) Ug = 1.2 cm/s (b) Ug = 3.8 cm/s (c) Ug = 7.7 cm/s.



(c)

Figure 2.5 The segment of the time series of photon count fluctuations recorded at Middle (Z/D = 1 and r/R = 0) for the liquid superficail velocity of 0.017 cm/sec and gas superficial velocity: (a) Ug = 1.2 cm/s (b) Ug = 3.8 cm/s (c) Ug = 7.7 cm/s(cont.).

We can see from the Figure 2.4 as the superficial gas velocity increases at constant superficial liquid velocity of 0.017 cm/sec the signal varies looks wavy and where the intensity of fluctuation of measured photon counts will change slightly from the range (60 – 80) to (60 – 100). The pulse flow regime in co-current upflow packed bed is characterized by increased in small bubbles inside the bed (Shah, 1979) that form as described earlier the gas rich followed by liquid rich flow pattern. Increasing in gas velocity, the voids between the bed particles will fill with both the gas – liquid which is called two phase pore flow (Raghavendra Rao et al., 2011). Thus, more randomly moving bubbles will intercept the path of γ-rays, and the signal looks unstable. At the middle section of the moving packed

bed as shown in Figure 2.5 the little expansion in the bed as the superficial gas velocity increased will introduce more and could be larger bubbles. Thus, appearance of peaks occurs as a succession of several ones. At higher superficial gas velocity (Ug) these bubbles will intercept the path of γ-rays result in increased the higher peaks in the photon count fluctuation (Nedeltchev et al., 2011).

2.3.2 Kolmogorov Entropy (KE). This approach of time series analyses is generally used to determine the level of disorder and non-linear characteristics in a dynamic system. Kolmogorov Entropy is a state space analysis which includes other parameters too such as the attractor reconstruction, the correlation dimension, and entropy analysis (Sasic et al., 2007; Van Ommen et al., 2011). The advantage of attractor comparison as a certain state space methods in comparison with frequency domain methods is that they are more sensitive to small changes in the initial state after differential time steps, and this feature can be used for, e.g., on-line monitoring (Van Ommen et al., 2011).

Many systems are chaotic whose behaviors appear random at the beginning but can be defined in phase space by plotting the long-term evolution of the system to form an attractor (Briens and Ellis, 2005). The hydrodynamics of gas – liquid – solid moving bed exhibit many features of chaotic dynamic systems, that are caused either by the flow of the phases through the bed and the voids of the fixed bed catalysts or by the frequent formation and breaking of bubbles. A chaotic system is a highly non-linear deterministic system that is usually characterized by its sensitive dependence on small changes in initial conditions (Gourich et al., 2006). Generally, the evolution of two initial states of a system will be completely different after some time, even they are started almost identical because the initial differences grow exponentially with time. The rate at which the disorder levels, that

in turn can be expressed in terms of quantities which is called Kolmogorov entropy (KE) represents the characteristics of the system disorder (Nedeltchev et al., 2003). Hence, the Kolmogorov entropy is used frequently to identify the flow regime, and to characterize the dynamics of the multiphase reactor. The Kolmogorov entropy value is a quantitative measure of the rate of information loss of the system dynamics and their disorders. It also quantifies the degree of unpredictability of the multiphase system. In most generic form, three different classes of systems can be distinguished depending on the rate at which these disorders increase (Nedeltchev et al., 2011):

- When the value of KE is zero which is limited cases for zero growth, the system is completely periodic (ordered) systems.
- When the value of KE is large (infinitely fast growth), the system is a purely random system making it impossible to predict the state of the system even after a differential time step, stochastic system.
- When the value of KE is small (finite), the system is in the case of more regular (periodic-like) behavior, chaotic system.

By means of the KE, some useful dynamic information within the time series can be extracted about the boundaries of the main hydrodynamic flow regimes. Thus in the present work, we use Kolmogorov Entropy which is calculated from the nonlinear chaos analysis to photon count time series. We will demonstrate its applicability to provide useful insights for the identification of the main flow regime boundaries in a moving bed as multiphase systems. The approach of Schouten; the maximum likelihood estimation of entropy (Schouten et al., 1994) is considered since it has been demonstrated of its successful implementation of pressure drop fluctuation signal to identify flow regime by

KE values (Nedeltchev et al., 2011). Therefore, in this work, we have applied the Schouten approach by programming it in a MATLAB program in our lab.

The maximum-likelihood estimator of the KE can be expressed as follows in Eq. (3.1).

$$KE = -f_s \, ln\left(1 - \frac{1}{b_m}\right) \tag{3.1}$$

Where:

b_m is the number of sequential pair of points on the attractor, and it's defined by:

$$b_m = \frac{1}{M}\sum_{i=1}^{M} b_i \tag{3.2}$$

f_s is the sampling frequency in (s^{-1}).

The attractor is basically set of values towards which the system evolves, and each data point is taken in this case. Hence, according to the time series when two points are move away that's mean a more chaotic system. The points in an experimental time series are measured at discrete, constant time intervals with a time step between two sampled data points that equals$\frac{1}{f_s}$. Following (Nedeltchev et al., 2007; Nedeltchev et al., 2006), the number of vector elements is set to 50, and the delay time is chosen to be unity.

$$AAD = \frac{1}{N}\sum_{i=1}^{N}|x_i - \bar{x}| \tag{3.3}$$

$\bar{x}$ is defined by eq. (3.4)

$$\bar{x} = \frac{1}{N}\sum_{i=1}^{N} x_i \tag{3.4}$$

In most of the cases, the number of vector elements in each state vector is equal to the embedding dimension. In this work, the maximum inter-points distance, (the so-called cut-off distance I_0) can be fixed at one, two or three times the average absolute deviation (AAD), when (I_0 = 3* AAD), as given by Eq. (3.3).

Researchers (Lin et al., 2001; Nedeltchev et al., 2007; Nedeltchev et al., 2011) used Kolmogorov entropy to identify the flow regime and the transition velocity in bubble column since it represents level of disorder. (Nedeltchev et al., 2012) identified the flow regime by calculating the Kolmogorov Entropy from the Gamma Ray Densitometry data for a fluidized bed. They have also found that nonlinear chaos analysis can be a useful tools and can be applied on the photon count time series data to identify the boundaries of the main flow regimes as well as their corresponding hydrodynamic behavior. The gamma ray densitometry data is a chaotic signal that measures in a time series.

2.3.3 Statistical Analysis. One of the simplest approach for the analysis of the time series is the statistical analysis in terms of calculating the standard deviation (σ) of the fluctuation as follows:

$$\sigma = \sqrt{\frac{\sum_{i=1}^{N}(x_i-\mu)^2}{N-1}}$$

Where (x_i) the data is point and (μ) is the mean and (N) is the total number of the data points. The transition from one flow regime to another has often been identified by the change in amplitude with operation conditions (Johnsson et al., 2000; Van Ommen et al., 2011).

3. RESULT AND DISCUSSION

3.1 FLOW REGIME IDENTIFICATION BY KOLMOGOROV ENTROPY (KE)

Figure 3.1 shows the KE values extracted from photon counts in moving bed reactor operated with air water as a function of Ug at the bottom section of the bed (Z/D = 0.3). We can see two local minimums in the KE curve. These minimum in the KE curve corresponding to the reorganization between two flow regimes, where the gas – liquid dispersion exhibits a self–organization step (Letzel et al., 1997; Nedeltchev et al., 2007). Hence, the peaks in the KE can be regarded as instability in the state of multiphase system, while the minimum KE can be regarded as stabilization states of the system (Nedeltchev et al., 2007). In other word, the peak in KE can be consider as a sensitive indicator of regime transition to other flow regime. In Figure 3.1, it sees higher KE value initial at very low flow rate and it can be due the phase maldistribution along the measured path of GRD. The value gradually decreases till Ug 1.2 cm/s this shows the flow is re-organizing or more organized in bubble regime itself. On increasing the Ug the KE value gradually increases it shows the flow is in transition towards pulse flow regime and on maximum value at Ug 3.8 cm/s the flow is transitioned to pulse flow regime. Then again the same trend repeats on increasing gas velocity, as the flow becomes more organized and then again transiting to other flow regime. In this case transition velocity from bubble flow to pulse flow is 3.8 cm/s, a similar result is reported by (Moreira and Freire, 2003b) for the transition region between bubble and transition I in their experiments in upflow mode. They mentioned that the interface between the bubble flow and transition I appeared at air flowrate close to 3.2 cm/s (0.04 kg/m^2 s), and it is similar to the result reported by (Colli-Serrano and Midoux, 2000). This transition region shows a little dependence on the water flowrate as they

explained. The flow regime transition is not a sharp condition rather than a range of conditions. (Iliuta and Thyrion, 1997) noticed visually a common transition region between the bubble and pulse flow regimes (7.6 < Ug < 10 cm/s) for air – water upflow with 3.3 mm porous particles.

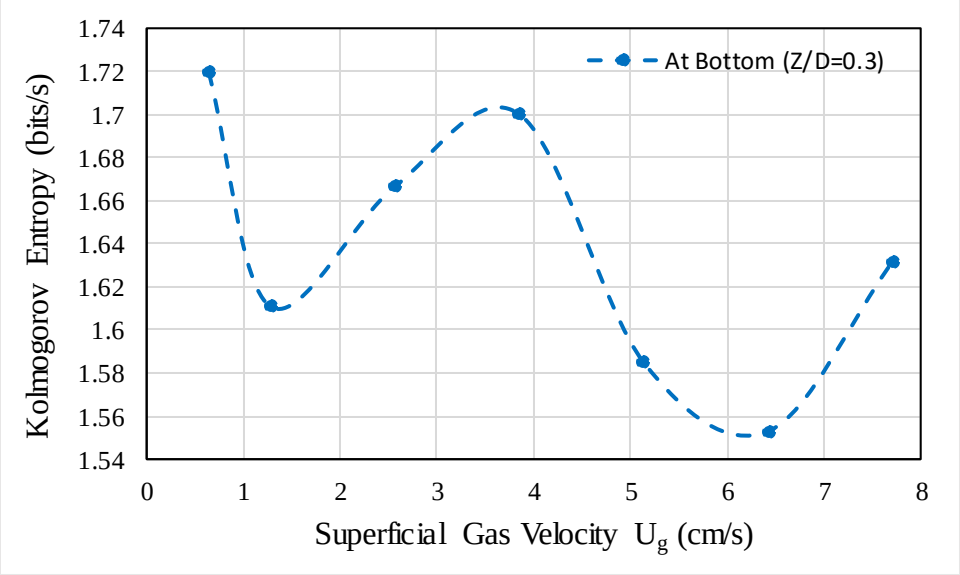


Figure 3.1 Kolmogorov entropy at the Bottom (Z/D = 0.3) of the moving packed bed column at center (r/R = 0).

Figure 3.2 below shows the KE values at the middle section of the moving packed bed and at the center region of the bed (r/R = 0). This region although behaves as packed bed in operational range is slightly expanded compared to the bottom part of the bed (Z/D=0.3). It shows clearly at Ug = 1.2 cm/s the first local maximum occurs, at these low gas flow rate conditions the pressure drop is less across the bed and the movement of gas and liquid is in disorder state due to channeling, and other forms of chaotic flow although it is in bubbly flow.

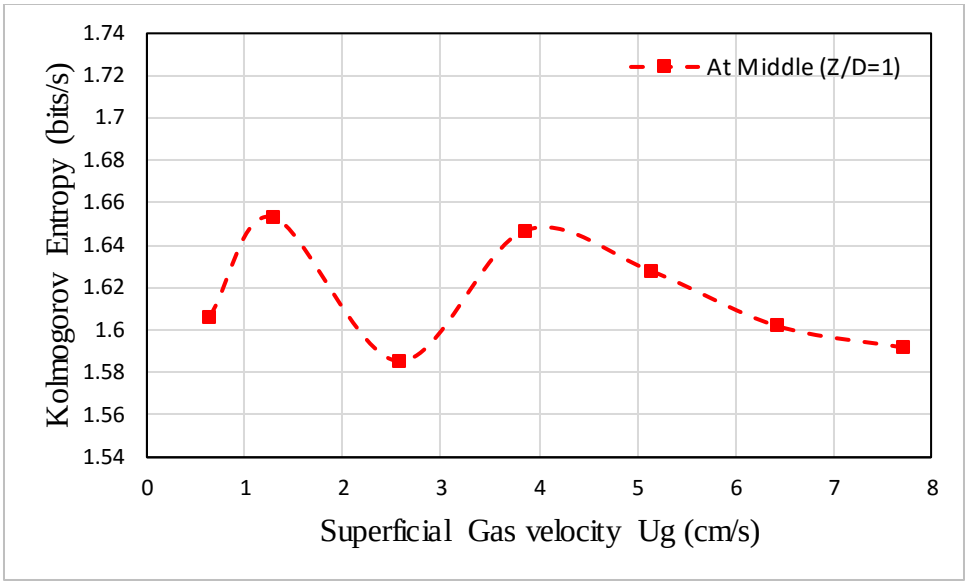
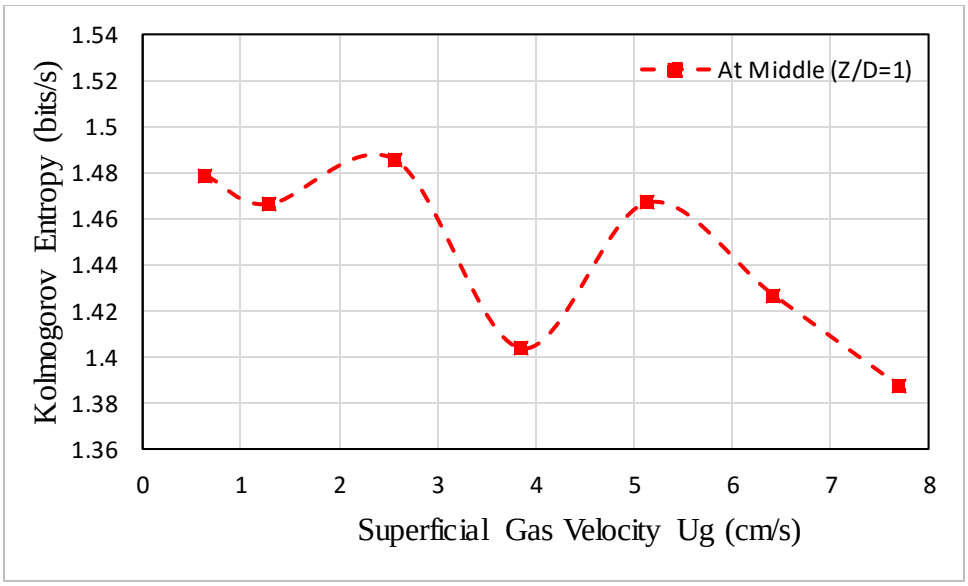
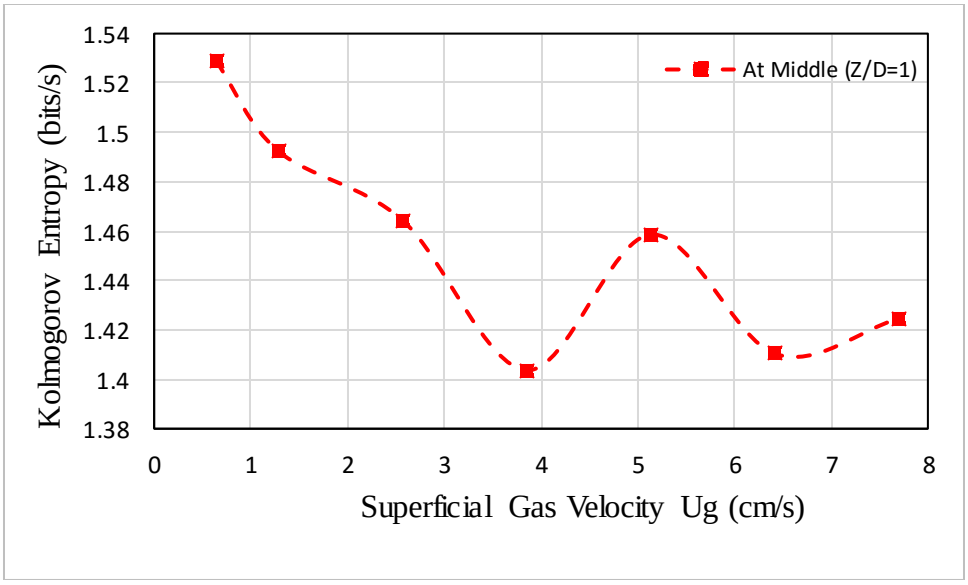


Figure 3.2 Kolmogorov entropy at the Middle (Z/D = 1) of the moving packed bed column at center (r/R = 0).

The same trend is repeated here also as seen in the Figure 3.1. At Ug 3.8 cm/s the onset of the pulse regime was discriminated, at this point the second local minimum occurs and this point of instability is due to regime change. When the pulse is entered the slug of liquid is followed by slug of gas and this flow structure is less disordered and hence KE reduces as gas flow rate is increased and it can be seen in Figure 3.2 after 3.8cm/sec increasing the gas velocity the KE decreases gradually as the flow become more organized and seems to be in fully developed region. This changing in the trend of flow regimes happened at increased in gas velocity 0.6-7.7 cm/s with constant liquid flowrate at 0.017 cm/s. (Raghavendra Rao et al., 2011) observed the bubble flow, pulse flow, and spray flow as the gas rate increases from low rates to high rates at a constant liquid flow rate in two phase up-flow packed bed. Since the range of velocity of gas (air) within a low to moderate we didn't face the spray flow, which happens at very high gas superficial velocity.

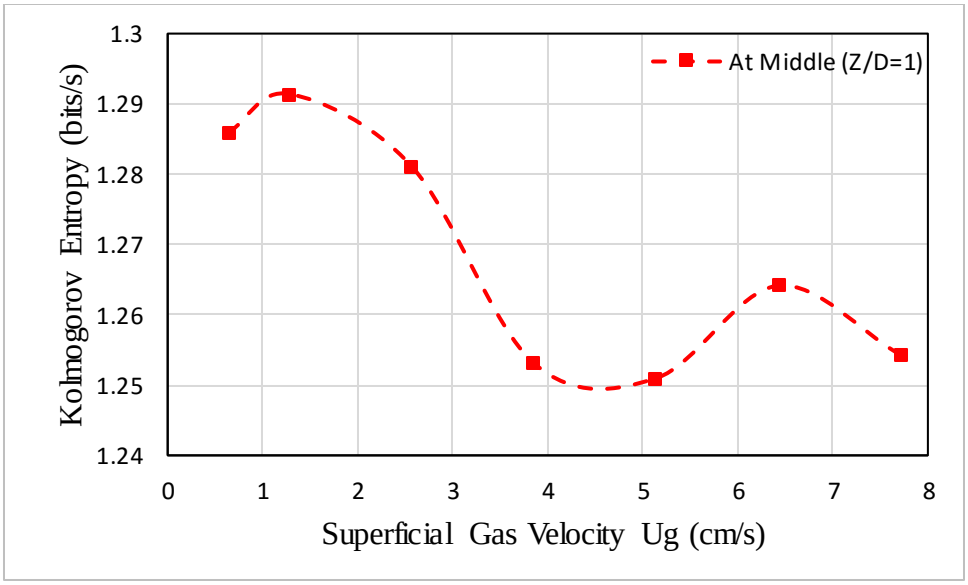


(a)

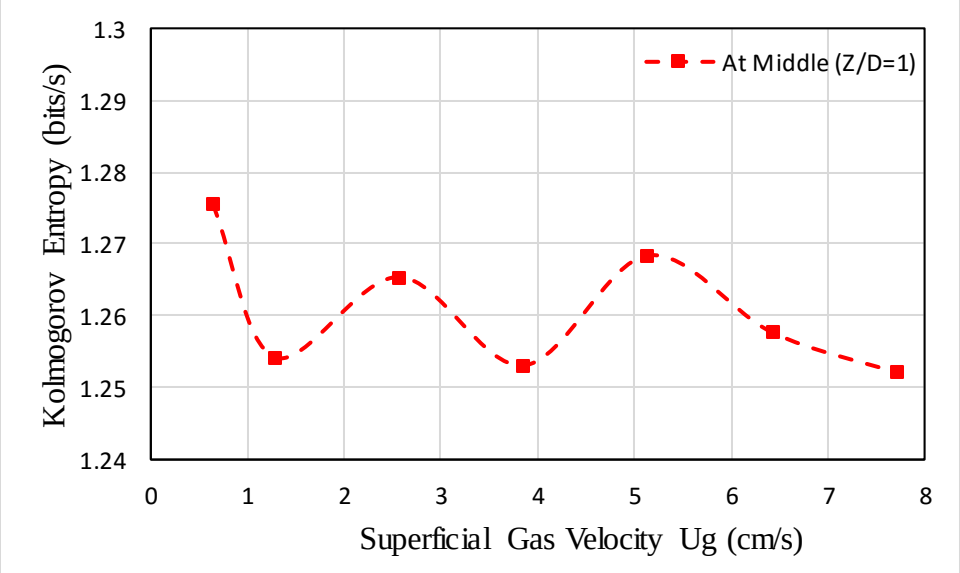


(b)

Figure 3.3 Kolmogorov entropy (KE) at the Middle (Z/D = 1) of the moving packed bed column: (a) at (r/R = 0.5), and (b) at (r/R = - 0.5).



(a)



(b)

Figure 3.4 Kolmogorov entropy (KE) at the Middle (Z/D = 1) of the moving packed bed column: (a) at (r/R = 0.9), and (b) at (r/R = - 0.9).

Figure 3.3 and Figure 3.4 represent the Kolmogorov Entropy as a function of superficial gas velocity Ug at the axial position (Z/D = 1) middle section of the moving packed bed and the radial position (r/R = ± 0.5) and (r/R = ± 0.9), from the center toward the wall region. We can see clearly that the values of the KE is lower than their values at the center of the bed. KE at the radial position (r/R = 0) with the average values = 1.6163, while the average values = 1.26 at the wall region of the column. (Muzen and Cassanello, 2007) obtained a low KE value from the time series at the wall measured in their experiments. They explained that these values corresponded to the existence of liquid slugs or waves at the wall region. The transition pattern is quite different among different locations of the bed it clearly shows how maldistributed the entire system is along both radial and axial directions. So the first local maximum represents the bubbly flow itself. but the high KE value is attributed to the chaotic nature arises due to channeling, bypassing etc.

3.2 FLOW REGIME IDENTIFICATION BY STATISTICAL ANALYSIS

At low gas superficial velocity, the curve of the standard deviation starts from high value and then decreases, this is because at low superficial gas velocity the disordered behavior of the system due to channeling, bypassing or other form of chaotic flow is prominent. The transition point can be identified from plotting the standard deviation of the signal, and its value varied with the superficial gas velocities as shown in Figure 3.5. The figure shows that the evolution of the standard deviation for two axial positions along the center of the bed. The plot of the standard deviation gives a less clear view of the transition as an increase in trend of standard deviation is observed at U_g = 3.8 cm/s. The slope of the standard deviation curve varied at this gas velocity, and this marked the onset

of transition region. As mentioned earlier a result reported by (Moreira and Freire, 2003b) for the transition region between bubble and transition I appeared at air flowrate close to 3.2 cm/s, and it is similar to the result reported by (Colli-Serrano and Midoux, 2000). (Raghavendra Rao et al., 2011) found that the bubble flow regime agreed with the bubble and transition I regime detected by (Moreira and Freire, 2003a). In deep pulsing regime the system is more ordered as seen from the KE findings. It is also visible from the standard deviation plot (Figure 3.5) that higher flow rate the standard deviation increase is in very small increment on increasing gas flow rate. In two phase upflow packed bed reactor, we may face a pulse flow regime at the bottom section of the packed bed which represents by the increase in small diameter gas bubbles as gas flowrate increased (Shah, 1979). (Varma et al., 1997) noticed by visual observation the pulses at the transition between the bubble flow and the pulse flow originate at the bottom of the packed column and move to the top of the packed column, and they also noticed the frequency of the pulses increasing with increase in gas rate. (Raghavendra Rao et al., 2011) explain that the pulse flow regime will be developed at low liquid flowrate by coalescing of the gas bubbles. Low liquid flow rate results in a low liquid velocity as passed through the voids of the bed. They explain that the transition line between the bubble and pulse flow regimes depends strongly on the gas flow rates. Where, the liquid velocity is lower than the gas velocity and can't remove the gas bubbles upwards through the bed particles. The photon counts fluctuations will increase as more gas bubbles intercept the gamma ray beam in the voids along the beam path. The density of the gas phase, in this case (air), is lower than the liquid phase (water). Therefore, the interaction between the gamma ray beam and the atoms of the gas phase (air) is lower than the interaction with the liquid phase (water).

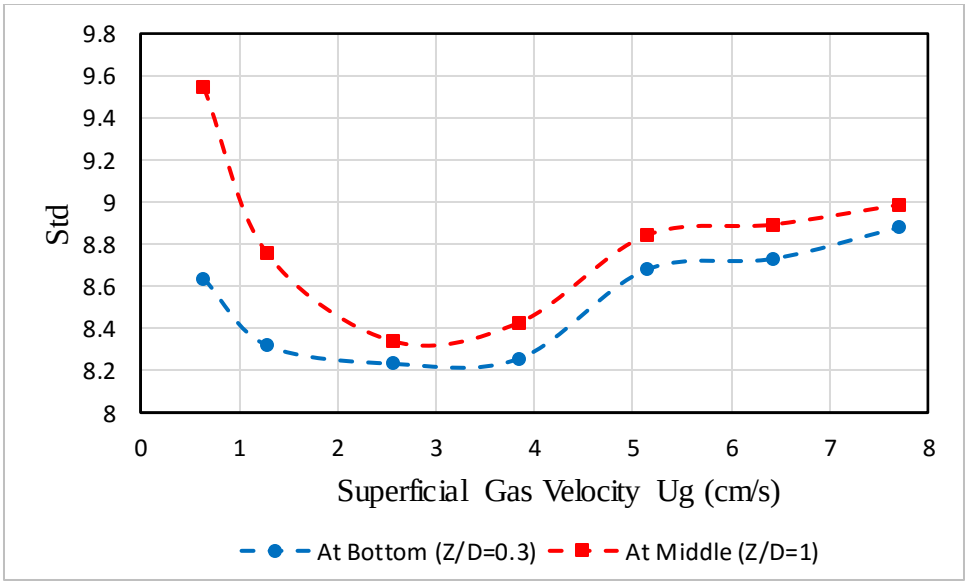


Figure 3.5 STD profile as a function of the superficial gas velocity U_g at the center (r/R = 0) of the moving packed bed column.

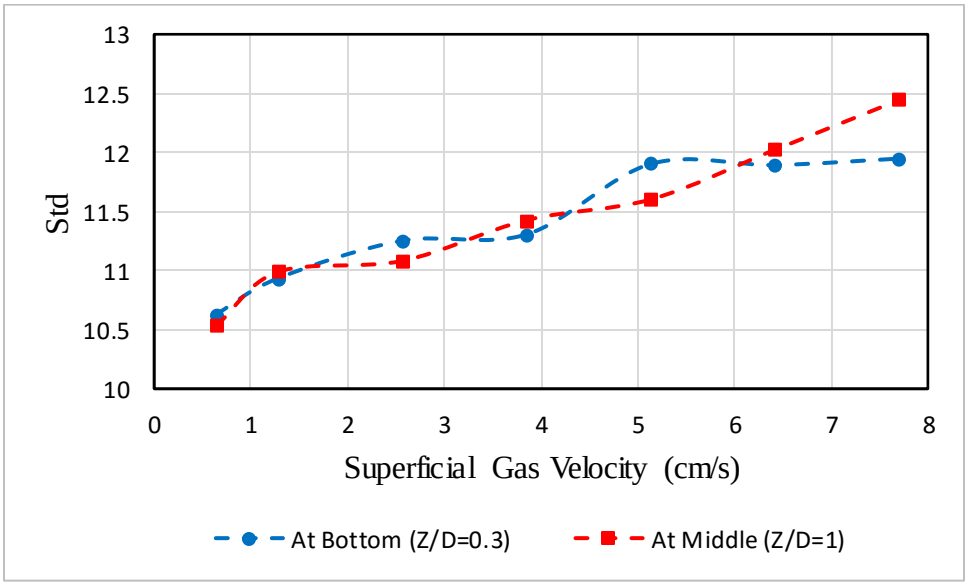


Figure 3.6 STD as a function of the superficial gas velocity U_g at (r/R = 0.5) right of moving packed bed column.

Figure 3.6 and Figure 3.7 show a noticeable change in the slop of the standard deviation curve around superficial gas velocity of 3.8 cm/s at the bottom (Z/D=0.3) where

the pulse flow regime starts. Based on the chaos analysis, transition from the bubble flow regime to transition flow occur at the same superficial gas velocity. As we can see, the value of the standard deviation is low at low superficial gas velocity. A larger change in the slop of the standard deviation curve observed after this velocity (3.8 cm/s). For their experiments in upflow packed bed, (Varma et al., 1997) noticed the transition between the regimes is not sharp and occurs at a small range in the gas and liquid flowrates.

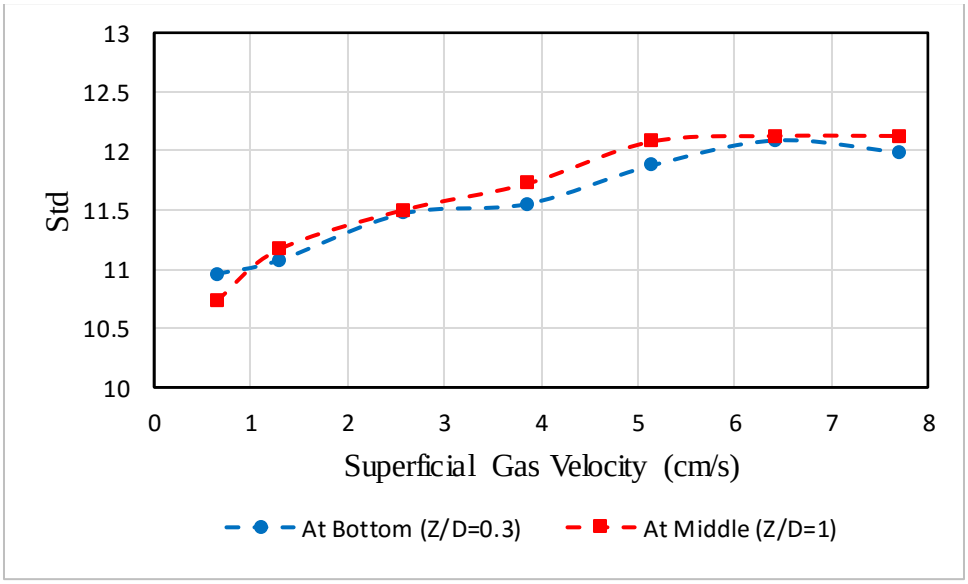


Figure 3.7 STD as a function of the superficial gas velocity Ug at (r/R = - 0.5) left of moving packed bed column.

As a summary, the curves of the standard deviation in Figure 3.6 and Figure 3.7, demonstrate that the deviation from the average value at each superficial gas velocity became larger and reached a relative stable level when the superficial gas velocity was greater than 3.8 cm/s. This behavior implies that the photon time series underwent large fluctuations as the gas velocity increased.

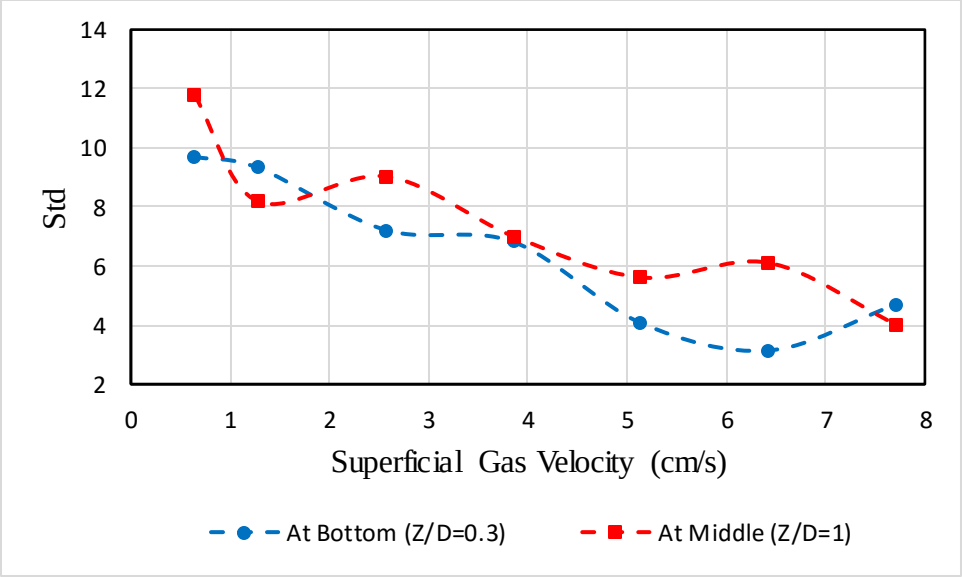


Figure 3.8 STD as a function of the superficial gas velocity Ug at (r/R = 0.9) right of moving packed bed column.

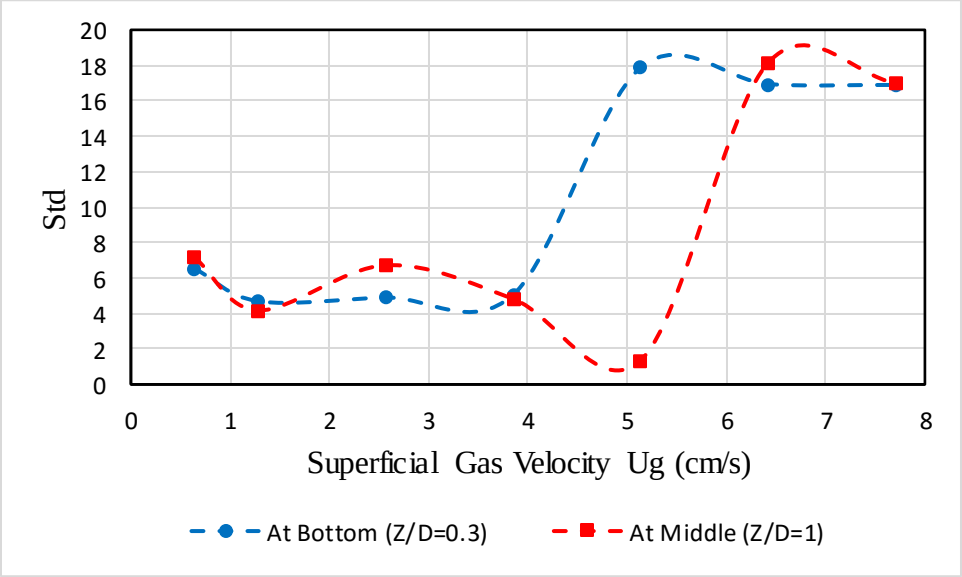


Figure 3.9 STD as a function of the superficial gas velocity Ug at (r/R = - 0.9) left of moving packed bed column.

The movement of liquid and gas bubbles along the bed is restricted by the solid catalyst and the pore dimension. That will affect several deterministic physical phenomena such as bubbles formation, passing, coalescence and breakup, since the column was packed randomly with catalyst particles. (Jo and Revankar, 2009) saw different flow behavior along different axial and radial positions in the packed bed, and explain the reason is due to the variety in the bubbles velocity as they moved through the pores of the packed bed. Figure 3.8 and Figure 3.9, show the standard deviation of photon counts at radial positions ($r/R = \pm 0.9$) respectively. We can see a different behavior for the curve of standard deviation which indicate a maldistribution of the two phases along the axial positions of the bed this is because at the wall the void fraction is high and approach unity where the flow structure varies due to least resistant to flow path (low pressure drop), hence in that region where both gas and liquid phase tend to move to flow there. (Jo and Revankar, 2009) explain that an increase and decrease in the local velocity of the gas – liquid phases flow can occur through narrow channels and pores along the path of the bed. The authors also explain there are stagnation points in the bed, where the bubble also can come to a complete stop. The static gas bubble will be isolated by a bridge of liquid, until its oscillation increased to release upwards or renewed by upcoming gas bubble. These behaviors in the column, in turn, will affect the gamma photon counts.

4. REMARKS

Bubble, transient, and pulse flow have been obtained at various axial and vertical positions by a noninvasive technique of gamma-ray densitometry in a co-current upflow moving packed bed column. The results were in a good agreement with the published data for upflow packed beds. The results demonstrated that the flow regimes of the upflow packed moving beds can be identified by the gamma ray densitometry technique by analyzing the time series of the photon counts which can be helpful in online monitoring in both laboratory and large scale industrial column with opaque wall. The photon counts were analyzed by both chaos and statistical approach. The boundary between the flow regimes were determined based on the trend of change in KE quantity and change in the slope of standard deviation curve. Kolmogorov Entropy (KE) showed that it can distinguish the transition region clearly than the statistical method based on the variation of standard deviation.

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SECTION

3. RECOMMENDATIONS

Although the current study provides useful information about moving packed bed reactor, many questions remain unanswered in topics of relevance to this work. Below are few recommendations for potential future research opportunities to yield a better understanding of the subject.

1. This work presents a deep insight on the liquid holdup, internal liquid holdup and average porosity in the packed bed section. However, it is limited to air water system under fixed liquid velocity to match the industrial operation conditions, as it assumed the perfect conditions. Therefore, it is important to adopt a study investigating these hydrodynamic parameters under various liquid superficial velocities, to assure the validity of the findings and results of the current work.
2. In future studies, it is necessary to investigate the effect of the distributor, and plenum sections on the two phase flow distribution and on other related hydrodynamic parameters.

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VITA

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