Gas Hydrodynamics and Mass Transfer in Low- and Medium-Consistency Pulp Suspensions in a Retention Tower

by

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ABSTRACT

In the pulp and paper industry, the interaction between the gas, liquid, and solid phases occurring in various unit operations is often not clearly understood. Such multiphase operations include flotation deinking (a separation process of paper fibres in the recycling process) as well as the delignification and bleaching operations in the kraft pulping process. Much of the design, operation, and optimization of such processing equipment are dependent upon past experience as well as trial-and-error methodologies.

Pulp fibre suspensions possess a complex and unique rheology. The unpredictability of the behaviour of pulp suspensions at any given mass concentration is due to the bonding between the fibres resulting in network formation (which depends on suspension consistency) with this interaction creating complexity in fluid flow in various unit operations.

This thesis describes the gas hydrodynamic behaviour and gas-liquid mass transfer characteristic in low- and medium-consistency pulp suspensions in batch operation. First, the hydrodynamic behaviour of the gas phase (air) in water and pulp suspensions having mass concentrations up to $C_m = 7\%$ is examined by visually observing and recording the bubble shape, size, and rise velocity in a rectangular channel. Results are obtained using a high-speed video camera. Second, the hydrodynamic behaviour is described in terms of the gas holdup along with axial and radial gas phase distributions in water and kraft pulp suspensions having mass concentrations between $C_m = 0.5$ and 9% in a batch-operated cylindrical bubble column. The gas holdup results are compared using three methods: the suspension height method, the pressure difference method, and the electrical resistance tomography (ERT) method. Finally, the volumetric gas-liquid mass transfer characteristic of air in water and kraft pulp suspensions having mass concentrations up to $C_m = 4\%$ is examined in the same bubble column in batch-operation using a dissolved oxygen probe.

Experimental results were comparable to that in literature for water and for pulp fibre suspensions having $C_m < 2\%$. The presence of fibres had a significant effect on the gas holdup and mass transfer characteristic with results providing insight on the limitations that exist in industrial pulp unit operations.

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NOMENCLATURE

Symbols	Definition, Units
a	gas-liquid interfacial area per unit dispersion volume, m^2/m^3
С	dissolved oxygen concentration, mg/L
ΔC	concentration gradient, kg/m ³
C_m	pulp mass consistency, %, or as a fraction in Equation 4.1 and 4.5
C_s	dissolved oxygen concentration given by sensor, mg/L
C^*	saturation concentration of oxygen in the liquid, mg/L
C_G	instantaneous concentrations of the transferring component in the
~	gas phase, kg/m ²
C_{Gi}	interfacial concentrations of the diffusing component in the gas $\frac{1}{2}$
C	pliase, kg/ill
C_L	linstantaneous concentrations of the transferring component in the
C	interfected constructions of the diffusing common on the liquid
C_{Li}	interfacial concentrations of the diffusing component in the inquid $n_{\text{hass}} = \frac{1}{2} \alpha / m^3$
D	phase, kg/m diffusivity m^2/c
D	ullusivity, III /s
D_c	diameter of volume equivalent subers m
a_e	Entropy number dimensionless
E_o	Entropy functional accoloration m/s^2
8 И	gravitational acceleration, m/s
П П	buddle height, chi height at position 1 m
Π_1 Π_2	height at position 2, m
112 H.	standy state as dispersed height suspension m
H	theoretical gas free height of suspension, m
I_{S}	transport flux kg/m^2s
J 12	mass transfor coefficient m/s
K Iz -	mass transfer coefficient m/s
KG k-	liquid film mass transfer coefficient m/s
nL k-a	x_{a}
k_u k	sensor time constant s ⁻¹
K _S	mass of fibres kg
m _f	mass of nulp suspension ka
mp m	mass of water kg
M_w	Morton number, dimensionless
	prossure drap between transducers in the presence of gas 1/De
ΔP	pressure drop between transducers in the presence of gas, kPa
ΔP_{o}	gas flow rate. L/min
$\mathcal{Q}_{\mathbf{p}}$	gas now rate, L/min Desmolds number, dimensionless
<i>Re</i>	keynolds number, dimensionless
l t	time, s
<i>l</i> ₁	time at position 1, s
<i>l</i> ₂	time at position 2, s

U U _b U _g	velocity of particle relative to continuous phase, m/s bubble rise velocity, m/s superficial gas velocity, m/s
Greek Letters	Definition, Units
δ	film thickness, m
δ_{G}	gas film thickness, m
δ_L	liquid film thickness, m
\mathcal{E}_{g}	gas holdup
μ	viscosity of continuous phase, Pa's
Δρ	absolute value of density difference between particle and continuous phase, kg/m ³
ρ	density of continuous phase, kg/m ³
$ ho_p$	density of pulp suspension, kg/m ³
σ	surface tension, N/m
σ_l	conductivity of the continuous phase, mS/cm
σ_2	conductivity of the dispersed phase, mS/cm
σ_{mc}	conductivity of the reconstructed measured conductivity by the ERT system, mS/cm
$ au_y$	yield stress, Pa

Acronyms

Definition

CEPA	Canadian Environmental Protection Agency
DP	degree of polymerization
ERT	electrical resistance tomography
FQA	Fibre Quality Analyzer
FXR	flash x-ray radiography
GDP	gross domestic product
HW	hardwood
NBSK	northern bleached softwood kraft
NPT	national pipe thread
PIV	particle image velocimetry
SGW	stone ground wood
SLPM	standard litres per minute
SW	softwood
TCDD	tetrachlorodibenzo-para-dioxin
TCDF	tetrachlorodibenzofuran

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DEDICATION

To Mom and Dad

CHAPTER 1

INTRODUCTION

1.1 Overview of Kraft Pulping Industry

1.1.1 Economic Aspects

The pulp and paper industry is one of the most important industries in Canada with 97 pulp mills, 170 paper mills, and 60 paperboard mills across Canada as of December 2005 (Industry Canada, 2006). Figure 1.1 presents the distribution of various pulp mills in Canada by province with British Columbia being the largest producer of chemical pulps.



Figure 1.1: Pulp, paper, and paperboard mills in Canada as of 2005

The pulp and paper industry employs over 56,000 Canadians directly with over 225,000 workers indirectly in the industry. Canada is the world's largest market pulp supplier, the fourth largest producer of pulp and paper products, and the world's number

one producer of newsprint with over 5.9 billion dollars in exports of wood pulp and other wood products and over 11 billion dollars in exports of newsprint and other paper and paperboard products in 2006 comprising 3% of Canada's gross domestic product (GDP) from the forestry sector (Industry Canada, 2006). Many communities in British Columbia and in other Canadian provinces rely on the forest industry sector as it is a large employer to many people especially in rural areas where it is the primary economic strong-hold of the community.

1.1.2 Pulping

The pulping process begins with the processing of the raw material consisting primarily of wood (90%) or other non-wood materials (10%) derived from plant matter for the production of pulp, paper and other paperboard products. The purpose of the pulping process is to liberate the fibres of the wood matrix to ultimately produce quality pulp that meets all customer requirements based on parameters such as cleanliness, brightness, and strength, in an environmentally responsible manner and at a minimum The two main mechanisms to liberate the fibres from the wood matrix are cost. mechanical and chemical pulping. In 2005, 93% of the world's market pulp consisted of chemical pulp, with the remainder, 7%, being mechanical pulp. This has been an increasing trend with most growth being of bleached softwood pulp (PPPC, 2007). In mechanical pulping, the wood is subjected to energy-intensive mechanical forces. The wood chips are shredded and de-fibred between rotating disks of a device called a refiner. Chemical pulping, on the other hand, separates the fibres of the wood matrix by degrading and dissolving the lignin (an undesirable high molecular weight amorphous material) that binds the cellulose fibres in the wood matrix through the application of chemicals. The presence of lignin in paper contributes to its degradation over time by breaking down and producing acids which weakens the fibres of the paper; therefore, the removal of lignin is the main objective of the entire pulping process and requires several unit operations in the pulping process to accomplish.

The dominant chemical pulping method is the kraft pulping process (or sulphate pulping). The kraft pulping process consists of cooking the wood chips in a solution of sodium hydroxide and sodium sulphide at high temperature and pressure for 60 - 180

minutes to dissolve the lignin of the wood matrix. This occurs in a unit called a digester. Once the fibres have been liberated and the lignin is dissolved to a satisfactory level, the pulp is then further processed in subsequent unit operations such as washing, screening, mixing, and further delignification. Up to this point, it is desirable to achieve the maximum delignification potential prior to subsequent bleaching stages in order to minimize the chemical charges required in the bleaching units. Figure 1.2 shows a simple schematic of a traditional kraft pulping cycle.



Figure 1.2: Schematic of kraft pulping cycle with oxygen delignification

In this thesis, the unit operation of interest in the kraft pulping process is the oxygen delignification tower. The quality of the pulp product following delignification is an important factor which will affect the efforts required in the subsequent bleaching stages. In order to achieve the best delignification in the tower to facilitate the

subsequent bleaching stages, the delignification must be optimized in such a way to achieve maximum lignin removal without damaging the fibres. In order to optimize the conditions in the oxygen delignification tower, the behaviour of the gas-liquid-fibre suspension must be understood. In particular, the distribution, hydrodynamics, and mass transfer characteristics of the three-phase suspension.

1.1.3 Oxygen Delignification and Bleaching

Early bleaching technologies relied heavily on chlorinated compounds for pulp bleaching since it functions as a good bleaching agent; however, the resulting effluent has a negative environmental impact. The environmental impact of the pulping and pulp bleaching processes has become of greater concern since the late 1980's, as strict effluent guidelines called for improvements in the pulping cycle and alternate bleaching practices for the responsible discharge of pulp mill effluent. For example, as of July 1, 1992, the Canadian Environmental Protection Agency (CEPA) has prohibited the release of 2, 3, 7, 8-tetrachlorodibenzo-para-dioxin (TCDD) and 2, 3, 7, 8-tetrachlorodibenzofuran (TCDF) in pulp and paper mill effluents (Environment Canada, 2007) and as a result, significant changes in bleaching technologies have been implemented. The industry has accepted the importance of being more environmentally conscious and responsible and in fact, 2002 reports on the pulp, paper and paperboard mills had operating expenses of 421.8 million dollars on environmental protection including 265.1 million dollars on pollution abatement and control processes and 69.2 million dollars on pollution prevention processes (Industry Canada, 2006).

Oxygen delignification is once such method that has environmental benefits. The process can be thought of as an extension to the cooking process that occurs in the digester, but more specifically, its objective is to remove the remaining lignin in the pulp suspension and to facilitate the next stage of the process, that is, bleaching. Molecular oxygen is applied at high pressure in an alkaline environment to remove lignin in pulp following the kraft pulping process and prior to the bleaching stages. Figure 1.3 illustrates a typical process. This process occurs in a retention tower with overall delignification attainable varying from mill to mill but typically between 35 - 50% delignification with higher delignification achieved with two-stage towers reported to be

as high as 65% (Bennington and Pineault, 1999). The resulting effluent, which is completely free of chlorine or chloride ion compounds, is recovered in the chemical recovery system. It is now a common practice in most pulping processes and has proven to be advantageous both economically and environmentally (San Fu, 2005; Patt *et al.*, 2005).



Figure 1.3: Flow diagram of a typical oxygen delignification stage

Although this method of delignification has no negative environmental impact, the delignification potential is limited due to concerns of fibre damage if extended periods of delignification were imposed. The concern is due to the natural creation of harmful free radical species which can attack cellulose and other carbohydrates. In addition, the presence of transition metals in the pulp can further catalyze the generation of these free radicals thereby decreasing the selectivity of the reaction. If the delignification can be improved in the oxygen delignification stage then this will be beneficial in the latter stages of the bleaching processes, lowering the demand of bleaching chemicals required and thereby reducing the impact on the waste treatment units.

The actual performance inside the tower is unknown. Gas hydrodynamics and uniformity of the pulp suspension need to be examined, as these will have an effect on the mass transfer and ultimately the delignification potential. An industrial mill survey conducted by Bennington and Pineault (1999) compared the process variables and operating parameters of several North American oxygen delignification systems in industry and no one specific cause of the delignification limitation could be identified. According to the data presented in the mill survey, each system was designed differently (height, diameter, high-, medium-, or low-consistency system) and operated under different conditions (consistency, entering kappa, temperature, pressure, pH, chemical charges, gas and pulp flow rates); therefore, it is thought that a modification to any one of these factors may affect the delignification achieved. This is observed by the variation in delignification efficiency achieved by the mills presented in the survey, 21 - 50%.

Although significant improvements in the gas-liquid mass transfer have been made by improving the mixer efficiency (Bennington et al., 1997; Rewatkar and Bennington, 2000), a performance evaluation of an oxygen delignification system suggested that since the majority of the delignification occurs in the retention tower (and not in the mixer where oxygen is first introduced to the pulp), that perhaps the mass transfer limitations in the tower may explain the delignification limitation and that the focus should turn towards the tower design and operating parameters rather than the mixer design (Van Heiningen, et al., 2003). Hence, an investigation into the hydrodynamics and mass transfer issues may explain the limitations in the achieved delignification potential. The difficulty with investigating this issue in industrial systems is that industrial oxygen delignification retention towers are large (aspect ratios (height to diameter ratio) between 6 and 16) pressurized stainless steel vessels making it virtually impossible to monitor what occurs inside the vessel without disturbing the operation. Issues concerning the uniformity of the pulp suspension, the hydrodynamics of the gasliquid suspension and mass transfer characteristics are of particular interest in order to gain an understanding of the reactor tower performance. Past work examining these issues has been conducted in low-to-medium consistency systems (Lindsay *et al.*, 1995; Reese et al., 1996; Pineault, 1999; Su and Heindel, 2003; Xie et al., 2003; Rewatkar and Bennington, 2004; Hol and Heindel, 2005).

1.2 Motivation and Objectives of Thesis

The purpose of this thesis is to describe the hydrodynamic behaviour and gasliquid mass transfer characteristics of the gas-liquid-fibre pulp suspension in low- and medium-consistency pulp suspensions (fibre mass concentration, $C_m = 0 - 9\%$) in a retention tower operating in batch mode. The operating parameters to investigate these phenomena include the superficial gas velocity, pulp type and fibre mass concentration (pulp consistency). The hydrodynamic behaviour including the bubble size, shape, velocity and location is examined in two types of geometric configurations, a rectangular bubble column and the more conventional cylindrical bubble column. The rectangular bubble column is used to study the bubble behaviour in pulp suspensions at various mass concentrations. This method has been used in literature (Krepper *et al.*, 2007; Zaruba *et al.*, 2005(a,b); Buwa and Ranade, 2005; Buwa and Ranade, 2004; Vandu *et al.*, 2004; Magaud *et al.*, 2001; Krishna and van Baten, 1999; Reese *et al.*, 1996) to visually observe the behaviour of bubbles rising in opaque suspensions since it is more difficult to accurately observe this behaviour in cylindrical columns.

There are many studies describing the hydrodynamic flow and mass transfer characteristics concerning multi-phase fluids; however, most of these reviews are concerned with batch flow operations with transient gas-phase flow (Letzel et al., 1999; Vandu and Krishna, 2003; Korpijarvi et al., 1999; Reese et al., 1996; Tecante and Choplin, 1993; Kawase et al., 1997; Patel et al., 2004), or in those cases with co-current flow of the liquid and gas phases, the material being investigated is one other than pulp suspension (Chaumat et al., 2004; Luo and Ghiaasiaan, 1997; Kikuchi et al., 1995). These studies have merit in certain applications in that it provides insight into the gas flow behaviour under specific conditions; however, they do not account for the actual behaviour of the pulp suspension (since a pulp suspension has a unique and complex rheology) occurring in a pulp retention tower for which we want to be able to characterize. Most relevant investigations to-date concerning gas-liquid hydrodynamics and mass transfer characteristics of pulp suspensions have been described by Pineault and Bennington (2002) (for gas hydrodynamics in pulp retention towers), Xie et al. (2003) (for gas hydrodynamics of pulp suspension flows), Lindsay, et al. (1995) (for hydrodynamic characteristics of pulp suspensions with batch and co-current flow configurations), Tang and Heindel (2005) (for gas-liquid-fibre flow in a co-current bubble column), Rewatkar and Bennington (2004) (for mass transfer characteristics in pulp retention towers), Rewatkar and Bennington (2000) and Bennington *et al.* (1997) (for gas-liquid mass transfer of pulp suspensions in mixers). However, further investigation is needed in order to describe both the hydrodynamics and mass transfer characteristics of pulp fibre suspensions in low- and medium-consistency oxygen delignification pulp retention towers for a wide range of operating parameters characteristic to industrial conditions. Susilo (2005) developed a model to predict the significance of gas-liquid mass transfer in pulp processing applications given the reaction parameters and operating conditions. Results showed that the delignification could be improved if modifications to the mixing and tower operating conditions were made. In order to determine what variables to modify, a clearer picture of the performance in the pulp retention tower is required.

It is important to investigate the hydrodynamic behaviour and mass transfer characteristics of pulp suspensions in co-current flow because industrial conditions dictate that the pulp suspension and gas phases are in motion at specific velocities. By understanding the flow regimes in the oxygen delignification tower as they behave in industry, as well as examining the volumetric mass transfer throughout the tower height, one can establish a better understanding of the degree of reactor performance and obtain a better understanding of the actual performance in the oxygen delignification tower. The specific objectives of this thesis are:

- To determine the shape, size, and rise velocity of air bubbles in water and kraft pulp suspensions having mass concentrations (pulp consistencies) between 1 and 7% in a rectangular vessel.
- 2) To describe the hydrodynamic behaviour, in terms of the gas holdup, and the radial and axial distribution of the gas phase in water and kraft pulp suspensions having mass concentrations between 0.5 and 9% in a batch-operated bubble column.

 To determine the volumetric gas-liquid mass transfer characteristics using air with water and kraft pulp suspensions having mass concentrations between 0.5 and 4% in a batch-operated bubble column.

To achieve the objectives, the following equipment is used for the rectangular vessel: a high-speed video camera, a high-intensity photographic lamp, and a diffuser, and for the cylindrical tower studies: an electrical resistance tomography (ERT) unit, pressure transducers, and a dissolved oxygen probe.

1.3 Scope of Thesis

First, an overview of the pulping process will be described, providing background information of the kraft pulping process including the oxygen delignification process in particular. Second, a presentation of more detailed background and a general literature review discussing previous contributions in the areas of hydrodynamics and mass transfer characteristics of multiphase fluid flow in general will be analysed and linked with the motivation for this thesis in particular. Also, the hydrodynamics and flow regimes concerning oxygen delignification towers is described including a discussion on the previous work done at UBC in the Pulp and Paper Centre concerning the hydrodynamics and mass transfer characteristics of pulp suspensions. Third, a description of the twodimensional experimental apparatus used for this study as well as a description of the technical methods and procedures carried out for the experimental investigations; this section also includes the results and discussion pertaining to the rectangular channel. Fourth, a description of the bubble column experimental apparatus, technical methods and procedures carried out, and includes the results and discussion linked with the rectangular vessel observations. Finally, conclusions and recommendations for future work are suggested.

CHAPTER 2

BACKGROUND AND LITERATURE REVIEW

2.1 Wood Fibre Composition, Properties, and Characteristics

The fibres used in the pulp and papermaking process are derived from two main categories of wood: softwoods and hardwoods. Softwoods, also known as coniferous trees, are generally larger compared to hardwoods, with longer fibres and wide lumens (void space in the fibre). Hardwoods, also known as deciduous trees, are shorter and have a thicker fibre wall compared to softwoods. The inherent physical and chemical properties of wood fibres have a strong influence on the papermaking potential and most other end-uses of papermaking products. The length of the fibre greatly affects the strength of the resulting pulp and paper product. On the other hand, fibre width and wall thickness affects fibre flexibility and its tendency to collapse in the paper production process. Fibres with thin walls collapse more readily and thus are more conformable, resulting in better bonding ability in a sheet structure, more dense, strong, and smooth sheets. In addition to the physical and chemical fibre properties, the pulp processing method used to extract the fibres also affects the quality of the end product. For example, mechanical pulping tends to produce shorter fibres due to the mechanical action which shortens the fibres compared to the chemical pulping method. Table 2.1 lists the fibre properties of various softwood and hardwood tree species.

Figure 2.1 shows the structure of the cell wall consisting of three layers: central lamina (middle lamina), primary wall, and secondary wall. The central lamina neighbours the primary wall of adjacent cells, and is comprised of pectic compounds, protein, and is lignin-rich. The primary wall is formed by microfibrils which are an irregular, net-like, interwoven pattern covering layer. The secondary wall is subdivided into three layers: outer layer, middle layer and inner layer and is responsible for much of the rigidity of the fibre and is made up of cellulose, hemicellulose, and lignin.

Species	Fibre Length (mm)	Fibre Diameter (µm)	Fibre Wall Thickness (µm)	Fibre Coarseness (mg/100m)
Softwoods				
Black Spruce	3.5	25 - 30	2 - 3	18
Douglas Fir	3.9	35 - 45	3 – 8	24
Loblolly Pine	3.6	35 - 45	4 - 11	18
Redwood	6.1	50 - 65	3.10	26.8
Splash Pine	4.6	35 - 45	3.8	18
Hardwoods				
Aspen	1.04	10 - 27	3.2	8.59
Birch	1.85	20-36	2 - 4	13.08
Oak	1.40	14 - 22	5 - 6	14.08
Red Gum	1.70	20 - 40		

Table 2.1: Fibre properties of various pulpwood species (Genco, 1996; Rydholm, 1965)



Figure 2.1: Structure of wood cell wall (PPRIS, 2008)

Wood is made up of carbohydrates (consisting of cellulose and hemicelluloses), lignin, and extractives. Cellulose is the most abundant compound in wood, constituting 40 to 50% of most wood species. It is a carbohydrate linear polymer derived from glucose with the chemical formula $(C_6H_{10}O_5)_n$ where *n* signifies the number of repeating units (monomers) or the degree of polymerization (DP). The DP varies with different sources of fibre and the treatment method (mechanical or chemical pulping for example) imposed to extract these fibres from the wood source. The DP typically ranges from 600 – 1500 and it is the DP that determines the quality of the fibre and its use in the papermaking process. Consequently, the higher the DP, the stronger the fibre due to the powerful associative forces (Smook, 1992). However, when the pulp undergoes bleaching, fragments of cellulose are created which are dissolved in solution, thereby decreasing overall pulp strength. In order to quantify the extent of cellulose degradation, the average length of the cellulose molecules are determined by dissolving the pulp in a solvent and measuring the viscosity of the solution. The measured viscosity is a function of the cellulose average chain length.

Hemicelluloses are also carbohydrates but are polymers of several different carbohydrates consisting of mainly hexoses and pentoses along with uronic acids which are linked non-uniformly. Hemicelluloses comprise 25 to 35% of wood. Also, they are branched, possess a lower molecular weight than cellulose, and are more easily degraded during chemical treatment than cellulose. Although they degrade more easily, hemicellulose does not contribute as much to the pulp strength as they do to the bonding property in paper.

Lignin is an extremely complex highly polymerized randomly linked amorphous compound consisting primarily of phenyl propane units and is the second most abundant organic compound on earth after cellulose. It is found throughout the cell wall of the wood but is concentrated mostly outside the cell walls and between each cell. It comprises 25 to 30% of typical softwoods and 20 to 25% of typical hardwoods. Lignin is often referred to as the "glue" that binds the fibres together as it is difficult to degrade but must be removed before manufacturing pulp and paper products and it must be done in such a way as to not damage the cellulose component for strength preservation. Figure 2.2 shows a model structure of lignin in a softwood.

Extractives constitute a small percentage of the fibre chemistry and may include resin acids, fatty acids, turpenoid compounds, and alcohols, for example, most being soluble in water or neutral organic solvents and thus easily removed from the wood matrix.



Figure 2.2: Structural model of spruce lignin (Genco, 1996)

The distribution of components in a fibre differs for each species and even differs for each individual tree within a species due to, for example, environmental factors. The average chemical compositions of the cell wall for softwoods and hardwoods are presented in Table 2.2.

Component	Softwood ^a (%)	Hardwood ^a (%)	
Carbohydrate	65 - 75	68 - 84	
Cellulose	40 - 45	38 - 49	
Hemicellulose	25 - 30	30 - 35	
Lignin	25 - 30	20 - 25	
Extractives	2-5	3 - 7	

Table 2.2: Average chemical composition of the main constituents in the cell wall of a typical softwood and hardwood (Genco, 1996)

^aValues are given as % woody dry solids

2.2 Kraft Pulping Process

The dominant chemical pulping process in industry today is the kraft or sulphate process. In 1884, C.F. Dahl, a German chemist, was credited with the first patent for the kraft process. Kraft pulp (*kraft* meaning strength in German) characterizes the stronger pulp produced when sodium sulphide is included in the pulping liquor, compared to the pulp obtained if sodium hydroxide alone is used, as in the original soda process. The alternative term, the sulphate process, is derived from the use of sodium sulphate, Na₂SO₄, as a makeup chemical in the recovery process (Biermann, 1993; Smook 1992).

The objective of the kraft pulping process is to remove the lignin in the wood matrix without degrading the cellulose component (the strength-characterizing component of the matrix).

The kraft pulping process makes use of sodium hydroxide and sodium sulphide at a pH greater than 12, temperatures between $160 - 180^{\circ}$ C, and at a pressure of 800 kPa for 0.5 - 3 hours to dissolve the lignin in the wood fibre. This takes place in a vessel called a digester. Lignin occurs naturally in wood along with cellulose and hemicellulose; however, because of its nature to degrade and its ability to yellow, it is undesirable in pulp and paper, and its removal must be maximized. Lignin removal is not an easy task since it is a complex molecule with various types of linkages. Not all the lignin is separated from the wood matrix in the digesters; therefore, further methods of lignin separation and removal are incorporated in the kraft pulping process by means of the bleaching stages which involves chemicals that target lignin removal over carbohydrate degradation as opposed to the chemicals used in the pulping process (Biermann, 1993). In the bleaching stages, one such stage is the oxygen delignification process, where the remainder of the lignin removal is maximized before being sent on to further bleaching stages.

2.3 Oxygen Delignification

2.3.1 History

Oxygen delignification technology can be dated back to the 1860's where a pair of researchers from the USA, Joy and Campbell, were the first to be granted a patent on the use of oxygen in conjunction with pulping with the method of passing heated air through agitated pulp suspensions. In 1915, Mueller, also from the USA, proposed a pressurized system with the addition of alkali. Further improvements were made during the mid 1950's, one of which focused on integrating the delignification aspect with cellulose-preserving steps (cellulose is the strength-characterizing agent; when cellulose is degraded, the pulp viscosity decreases, and hence, the strength decreases). However, not until the 1960's has it been in common practice as a commercially feasible process in pulping process systems in France, Sweden, Japan, as well as South Africa, and has more recently been implemented in North America (Aker Kvaerner, 2005).

The interest in the application of oxygen in pulp delignification was realized due to the negative environmental impact that conventional bleaching operations had on the environment since conventional bleaching makes use of chlorinated compounds. Another incentive for the application of oxygen delignification process in the pulping process was the need for making the production process more cost-effective. Although the conventional bleaching methods were very selective towards lignin, the high cost of the bleaching chemicals as well as the installation and maintenance of effluent treatment systems were a disadvantage. Hence, the application of oxygen for pulp delignification as well as in bleaching operations was an attractive option because not only did it substantially reduce the pollution problems by reducing BOD (biochemical oxygen demand), colour, and chlorinated organic compounds in the effluent, but also, reduced the cost of expensive bleaching chemicals (Air Liquide, 2005; Dence and Reeve, 1996). However, the lack of lignin selectivity, as well as doubts about the negative effects on the strength of the pulp (the pulp is degraded due to the application of oxygen), was of concern.

In the mid 1960's, a group of researchers from France had demonstrated the positive effect of the addition of magnesium ions derived from magnesium salts such as

MgSO₄ and MgCO₃ to the pulp suspension which improved selectivity as well as controlled cellulose degradation, a parameter which characterizes pulp strength (Moe and Ragauskas, 1999). Hence, the implementations of full-scale mill trials were necessary to prove the technology viable. Although this technique proved to be a viable solution to the pollution problem, oxygen delignification was not widely accepted due to high investment costs, as well as the extensive formation of carbon monoxide which would accumulate at the top of the tower and cause puffs and/or explosions in high consistency systems. In order to prove that an oxygen delignification system was a viable solution in industry, process optimization was required, one of which, was the application of medium-consistency equipment which will be discussed in more detail later on, that allowed for more flexibility of process options (Aker Kvaerner, 2005).

2.3.2 Process Description

Oxygen delignification can be thought of as an extension to the cooking process that occurs in the digester, but more specifically, its objective is to remove as much of the remaining lignin in the pulp suspension and to facilitate the next stage of the process, i.e., bleaching. Oxygen delignification is now a common practice in most pulping processes and has proven to be advantageous both economically and environmentally (San Fu, 2005; Patt, Kordsachia, Suttinger *et al.*, 2005).

The oxygen delignification process in the context of the entire pulping process occurs following the kraft cook and prior to the bleaching processes. Please refer to Figure 2.3 for a schematic diagram of the entire pulping process. Following the kraft cook, there is still a significant quantity of lignin remaining in the fibre. The residual lignin content after kraft pulping is 2 - 5% by mass and is responsible for the dark colour of the unbleached pulp.

Oxygen delignification (or bleaching) is the application of molecular oxygen at high pressure in an alkaline environment to remove lignin in pulp following the kraft pulping process and prior to the bleaching stages. Its caustic effluent can then be added to the black liquor and processed through the recovery cycle via the brown stock washing system to recover valuable chemicals which are recycled to the pulping process and thus allows for further system closure of the pulping and bleaching processes. Oxygen delignification has grown remarkably as a result of continuing efforts by the industry to improve effluent quality. The effluent resulting from oxygen delignification is completely free of chlorine or chloride ion compounds and hence it can be fully recycled into the cooking waste liquor recovery system, which is now a standard practice in the pulping industry.


Figure 2.3: Schematic of a pulp mill. a) steaming drum; b) high-pressure valve to the continuous digester;
c) continuous digester for kraft and bisulfite pulping; d) waste liquor outlet; e) digestion liquor inlet; f) blow tank; g) filter for pulp washing; h₁) coarse screening; h₂) fine screening; i) filter thickener; j) chlorine bleaching tower; k) bleaching tower for alkali treatment; l) bleaching tower for hypochlorite treatment; m) bleaching tower for chlorine dioxide treatment; n) wash filters; o) mixers; p) storage tanks; q₁) coarse postscreening; q₂) fine postscreening; r) machine chest; s) dewatering machine; t) tunnel dryer; u) sheet cutter (Patt *et al.*, 2005)

2.3.3 Process Conditions and Operating Parameters

Two types of oxygen delignification systems that are in common use are: high consistency systems, containing 25 - 30% fibre concentration in pulp, and medium consistency systems, containing 10 - 14% fibre concentration in pulp. Medium consistency systems offer an advantage over high consistency systems in that the capital cost is lower because of the expensive dewatering equipment that would be required for high consistency systems. However, higher operating efficiencies with the high consistency systems are what attract some mills to these systems (Dence and Reeve, 1996).

The degree of delignification is commonly between 40 - 50%; however, greater degrees of delignification may be attained with two-stage systems which approach 65% delignification efficiencies. A mill survey conducted by Pineault and Bennington (1999) showed that the average delignification for softwoods was 47.5% with a range between 28% and 67%, and the average for hardwoods was 42% with a range between 29% and 55%. Typically, above 50% delignification, pulp strength can become compromised due to the relatively low selectivity of oxygen in the delignification process; however, the addition of magnesium salt as a cellulose protector has been investigated and aids in this manner as well as the introduction of two-stage systems (Genco, 1996; Moe and Ragauskas, 1999). Table 2.3 shows the typical operating conditions for medium and high consistency systems.

	System Condition			
Parameter	Medium Consistency	High Consistency		
Consistency, %	10 - 14	25-30		
NaOH Charge, % [*]	3	2.5		
Retention time, minutes	60	30		
Temperature, °C	90	110		
Pressure, kPa	500	500		
Oxygen consumption, %*	2-3	2-3		

Table 2.3: Typical operating conditions for medium and high consistency oxygen delignification systems

^{*}Percentage on oven-dry pulp

2.3.4 Medium Consistency Systems

There are several configurations which are available when employing medium consistency oxygen delignification systems. These include single stage, two-stage, and the more recent "mini" oxygen delignification systems. With a two-stage system, delignification efficiencies may approach 65%, which will greatly be of benefit in subsequent bleaching processes since this will greatly reduce the bleaching charge required. The more recent "mini" oxygen delignification systems are designed to accomplish the same delignification requirements as a single or two-stage system; however, at a significantly lower capital cost than the conventional process (San Fu).

Medium consistency oxygen delignification is carried out at a temperature of $85 - 110^{\circ}$ C with 60 minutes retention time. Brown stock is first washed to minimize carry over of black liquor solids that compete with lignin for reaction with oxygen. The pulp is then charged with 2 - 3 wt% NaOH as well as magnesium protector and preheated in a low-pressure steam mixer and then pumped through gas mixers which allows for sufficient dispersion of oxygen as small bubbles in the pulp suspension. The pulp is then sent to an upflow pressurized reaction tower so that the oxygen delignification reaction may reach completion. The stock is continuously discharged at the top of the tower, depressurized in a blow tank, and then sent to the washers. Approximately 2.5% oxygen is consumed based on oven-dry pulp and 40 - 45% reduction in lignin content may be achieved (Genco, 1996).

Figure 2.4 shows a process flow diagram of a conventional medium consistency oxygen delignification system. Medium consistency systems consume more oxygen and alkali, produces a slightly lower degree of delignification; however, they have the potential to produce a higher degree of delignification with the implementation of two-stage medium consistency systems, as well as the most recent "mini" oxygen delignification system. In the 1970's and 1980's, high consistency systems were mostly present for oxygen delignification; however, with the lower capital cost and inherently safer operation provided by medium consistency systems, they are mostly present in industry today. (Smook, 1992; Dence and Reeve, 1996; Genco, 1996).



Figure 2.4: Medium consistency system process flow diagram (Genco, 1996)

The medium consistency oxygen delignification reactor is the focus of this thesis. The volumetric mass transfer coefficient is a key parameter in determining reactor performance. In addition to the mass transfer parameter, the hydrodynamics and flow behaviour of the three-phase suspension (gas, liquid, and pulp fibre) is also of importance. If one were able to determine the distribution of mass transfer and flow characteristics throughout the column height, then one can gain a better understanding of the performance inside the tower.

2.4 Hydrodynamics of Pulp Fibre Suspensions

2.4.1 Rheology

Rheology is the study of the relation between momentum transfer and the structure of the flow field resulting in deformation and flow of matter under an applied Pulp suspensions have a very unique and complex rheology such that the stress. rheological properties (viscosity, surface tension) are not widely reported in literature. Pulp suspension flow and its rheological properties are most commonly characterized by non-Newtonian phenomena, a phenomena that exists when there is a non-linear dependence between the shear stress and shear rate of deformation. The behaviour of the pulp suspension is dependent on many factors such as: density, viscosity, concentration, fibre dimensions, fibre-fibre interactions, presence of other matter in the suspension, as well as factors that are imposed on the suspension such as temperature, shear forces, and turbulence, for example. When the fibre material is subjected to mechanical and chemical treatment, the fibre properties gradually change. Mechanical treatment may damage the structure of the cell wall and geometrical properties such as the fibre diameter, length, and fibre wall thickness thereby reducing the overall stiffness of the suspension and ultimately resulting in a weaker fibre suspension.

The degree of interaction between fibres in a flowing pulp suspension depends largely on the concentration of fibres present in the suspension. Fibre-fibre interactions lead to network formation (flocs of fibre bundles) that possesses network strength. The yield stress which characterizes the network strength must be overcome in order for the suspension to flow and is achieved by shear forces larger than the yield stress. In pulp fibres suspensions, the ability of the suspension to flow depends largely on the consistency of the suspension. At low mass concentrations ($C_m \leq 1\%$), the suspension flows with relative ease; however, as the consistency increases, increasing fibre-fibre interactions leading to network strength inhibits the suspension's ability to flow with ease.

Experimental studies of the yield stress of fibre suspensions have been examined previously (Gullichsen and Harkonen, 1981; Bennington *et al.*, 1990; Wikstrom and Rasmuson, 1998; Pettersson and Rasmuson, 2004). Bennington *et al.* (1990) determined

the yield stress for commercial wood pulp suspensions and synthetic fibre suspensions of low and medium mass concentration with measurements carried out in a rotary viscometer and in a concentric rotary shear tester. Results showed that not only did the yield stress depend on the volume of the suspension occupied by the fibres (mass concentration), but also that the properties of the fibres themselves were a contributing factor at constant consistency.

The yield stress was determined experimentally at room temperature (T = 20 °C) and pressure (P = 1 atm) for softwood and hardwood kraft pulps (Domtar, Inc., QC). Figure 2.5 presents the yield stress of both softwood and hardwood pulps as a function of consistency. The experimental results were correlated using an equation of the form:

$$\tau_y = a C_m^{\ b} \tag{2.1}$$

where *a* and *b* are constants specific to a fibre type and C_m is the pulp mass concentration defined in equation 2.2, with m_f as the mass of fibres and m_w as the mass of water, and expressed as a percent.

$$C_m = \frac{m_f}{m_f + m_w} * 100\%$$
(2.2)

The yield stress for softwood pulps is described by $\tau_y = 12.8C_m^{2.03}$ and hardwood pulps by $\tau_y = 1.23C_m^{3.00}$, and are specific to the pulps used in this study. Both pulps result in an increasing yield stress with increasing fibre mass concentration but the yield stress for the softwood pulp increases at a much faster rate than the hardwood pulp.



Figure 2.5: Yield stress as a function of consistency for softwood and hardwood kraft pulps

The network strength for softwood pulps is greater than that for hardwood pulps resulting in a greater stress required to break the network and can be explained due to the characteristics of the fibres themselves. The interaction of fibres and bonding forces in a pulp suspension causes the formation of fibre networks. These fibre networks possess a structure and a strength and will hydrodynamically behave differently depending on the type of fibre and concentration of fibres, for example. The consistency (or mass concentration) of the pulp suspension is an important factor because as the suspension consistency increases, so too does the degree fibre-fibre interaction as depicted in Figure 2.5. For example, pulp suspensions with low consistencies, $C_m = 0 - 4\%$, possess fewer fibre-fibre interactions than pulp suspensions with medium consistencies, $C_m = 8 - 16\%$, and are characterized by flocs of fibres in water as opposed to aggregate fibre structures behaving as separate entities in the pulp suspension (Dence and Reeve, 1996; Lindsay et al., 1995; Viswanathan, 1986). The addition of a third phase, gas, complicates the situation. Flocs of fibres can trap bubbles thereby preventing their rise through the suspension. Bubbles are forced to either bypass the network of flocs or coalesce into

larger bubbles with enough buoyant force to break through the network of fibres (Pelton and Piette, 1992). In stagnant pulp suspensions, bubbles are most likely to flow through channels with the lower resistance, as will be described later in Chapter 3.

It is important to understand the flow behaviour and rheological properties of pulp fibre suspensions because the design of pulp processing equipment will depend on it. For example, the design of a mixer in the oxygen delignification process will require that the gas is sufficiently dispersed in the pulp suspension to uniformly introduce the stock and avoid channelling and non-uniformities in the upflow towers. In the bleaching stages, the ability of the bleaching agent to penetrate the lignin in the fibre structure has a lot to do with the nature of the suspension. For example, the formation of fibre flocs at certain pulp mass concentrations may cause phase segregation resulting in non-uniform delignification.

2.4.2 Pulp Suspension Flow Regimes

The flow of a fibre suspension is influenced by:

- 1. The type of fibre and its inherent properties such as the length, diameter, cell wall width, density, flexibility, uniformity, surface structure.
- 2. The presence and interaction with other suspended matter such as air bubbles, fines, additives.
- 3. The forces that act on the suspension itself such as pressure, temperature, shear, and turbulence.
- 4. The environment that houses the suspension such as the vessel geometry and vessel properties (roughness).

The flow behaviour in a bubble column (a cylindrical reactor with a gas distributor at the bottom) is classified according to regimes maintained by the superficial gas velocity employed in the column. Three types of flow regimes commonly exist in bubble columns: homogeneous (bubbly flow) regime, heterogeneous (churn-turbulent) regime, and the slug flow regime (Godbole and Shah, 1986; Kantarci *et al.*, 2005; Zehner

and Kraume, 2005). Table 2.4 describes each flow regime and Figure 2.6 presents the regimes schematically.

Flow Regime	Bubble Size	Flow Description	Bubble Interaction	
Homogeneous	Small	Uniformly distributed	No bubble	
bubbly flow		across column cross-	coalescence or break-	
		section	up	
Heterogeneous or	Large and	Large bubbles rise in plug	Unstable bubble	
churn-turbulent flow	small	flow, small bubbles are	coalescence and	
		backmixed in pulp	break-up, resulting in	
		suspension	varied bubble sizes	
Slug flow	Large	Bubbles fill entire cross-	Bubble slugs	
		section as they move	resulting from bubble	
		upwards	coalescence	

Table 2.4: Flow behaviour in bubble columns



Figure 2.6: Schematic of bubble column flow regimes (Kantarci et al., 2005)

Parameters that affect column performance include column dimensions, gas distributor design, fluid properties, and operating conditions such as gas and pulp suspension flow rates, temperature, and pressure. The key parameters used to quantify the performance include the flow regime, gas hold-up, bubble size and distribution, and

bubble rise velocity. The gas hydrodynamics can change dramatically depending on the concentration and physical properties of the solid phase. For example, the presence of a higher concentration pulp suspension can increase bubble coalescence since the solids impede the flow of gas through the tower, trapping the gas bubbles and causing them to coalesce with other bubbles, hence, resulting in larger bubbles, a greater gas rise velocity, and decreased gas hold-up in the tower (Heindel, 2005; Pineault and Bennington, 2002).

The relationship between the superficial gas velocity and column diameter on the resultant flow regime is depicted in Figure 2.7 where the transition region is affected by factors such as those listed previously (distributor design, fluid properties, etc.).



Figure 2.7: Flow regimes in bubble columns (Zehner and Kraume, 2005)

The flow regimes can be extended to the gas holdup parameter (also called the void fraction). Gas holdup is a dimensionless parameter that characterizes the gas presence in a vessel. It is defined as the volume fraction of the gas phase occupied by gas bubbles. Similarly, liquid holdup and solid holdup define the volume fraction of liquid and solid phases, respectively. Further discussion on the gas holdup in this thesis is provided in Chapter 4.

The flow regimes in pulp suspensions have been studied previously (Lindsay *et al.*, 1995; Reese *et al.*, 1996; Su and Heindel, 2003; Xie *et al.*, 2003; Hol and Heindel, 2005).

Lindsay et al. (1995) conducted experimental studies on the hydrodynamic characteristics of three-phase slurry column with water, pulp fibres, and air. Two configurations were examined in a transparent 1.5 m long and 13 cm internal diameter column, one for quiescent liquid (batch operation) (with gas superficial velocities up to 4.2 cm/s) and one for co-current air and liquid flow (with suspension superficial velocities between 2.5 and 7.5 cm/s). Visual observations of the flow pattern and bubble behaviour in the column were limited to regions near the vessel wall. Bubbly flow regime was observed in two-phase water and air systems and with three-phase systems with low pulp mass concentrations and low superficial gas velocities. However, with higher gas superficial velocities, evidence of bubble coalescence was confirmed with the resultant lower gas holdup values, and the flow regime developed into a heterogeneous churn-turbulent flow pattern. In the quiescent flow configuration, the macroscopic flow patterns were found to be significantly different than those observed in pure water or nonfibrous three-phase systems. With 1% and 2% pulp mass concentrations studied, fibre flocculation and network formations affected the flow of the gas phase resulting in bubble coalescence and channelling phenomena. Similar results were obtained for the co-current configuration; however, the bulk flow of the liquid phase caused a decrease in bubble coalescence when the liquid superficial velocity was greater than that of the gas superficial velocity. The flow of the liquid phase carried away with it the gas bubbles before bubble coalescence could occur.

Reese *et al.* (1996) studied the hydrodynamic characteristics of three-phase slurry system with air, water and pulp fibres at mass concentrations between 0.1 and 1%. The hydrodynamic behaviour was studied in a batch configuration and is comparable to results presented by Lindsay *et al.* (1995). At low pulp mass concentrations (0.1 to 0.25%), the pulp is uniformly distributed throughout the entire column at all superficial gas velocities and the bubbly flow regime is present. This was also reported by Su and Heindel (2003). With higher pulp mass concentrations (0.75 to 1.0%) and higher superficial gas velocities, pulp fibres begin to form flocs, leading to a non-uniform fibre

distribution in the column and the flow regime transitions to one with bubble coalescence. Su and Heindel (2003) reported that distinct fibre networks were formed at pulp mass concentrations greater than 0.6% and classified the flow regime as heterogeneous for all gas superficial velocities.

Xie *et al.* (2003) studied the hydrodynamic co-current flow characteristics of pulp suspensions with mass concentrations between 0 and 1.5% in a vertical cylindrical column 1.80 m in height and 5.08 cm in diameter. They identified, visually as well as by flash X-ray photography, six distinct flow regimes: dispersed bubbly flow, layered bubbly flow, (incipient plug and) plug flow, churn-turbulent flow, and slug flow, and concluded that the flow regimes were dependent on consistency. Figure 2.8 schematically depicts each flow regime with increasing gas superficial velocities, respectively, where the arrows represent the apparent motion of the gas bubbles.



Figure 2.8: Schematic of flow regimes, (a) dispersed bubbly flow, (b) layered bubbly flow, (c) incipient plug flow, (d) plug flow, (e) churn-turbulent flow, and (f) slug flow (Xie *et al.*, 2003)

Flow regime maps in air and water mixtures as well as pulp suspensions detailing the regime as a function of both the superficial gas and liquid velocities were also described and illustrated.

The performance of a reactor is dictated to a great extent by bubble characteristics such as bubble size, bubble rise velocity, and wake phenomena. Mixing in a reactor is governed by the bubble characteristics: gas holdup, gas residence time, and interfacial area, an important aspect in the mass transfer performance.

2.5 Mass Transfer

Mass transfer is the transport of atoms or molecules from one spatial region to Mass transfer occurs when there is a concentration difference in random another. molecular motion. The transfer of mass is initiated through this concentration difference and as a result, the transfer of mass occurs from regions of high concentration to regions of low concentration. The amount of mass that is transferred may be quantified through the calculation and application of mass transfer coefficients. The rate at which mass transfer occurs depends mostly on factors that affect the interfacial area, a. Factors that may influence the gas-liquid mass transfer include temperature, pressure, viscosity, density, surface tension, surfactants, solids concentration, aeration velocity, and pH, to name a few. Figure 2.9 presents a general depiction of the dissolved oxygen concentration profile near the gas-liquid interface where C_G and C_L are the instantaneous concentrations of the transferring component in the gas and liquid phase, respectively, C_{Gi} and C_{Li} are the interfacial concentrations of the diffusing component in the gas and liquid phase, respectively, δ_G and δ_L are the gas and liquid film thickness, respectively.



Figure 2.9: Dissolved oxygen concentration profile near gas-liquid interface (Chisti, 2002)

The mass transfer is governed by diffusion assuming that the gas phase passes through stagnant gas and liquid films and the resistance to transfer is localized within the films with no resistance in the gas-liquid interface. At steady state, linear concentration profiles exist in the films and the transport flux, J, (rate of transfer per unit crosssectional area) of the diffusing species can be related to the concentration gradient, ΔC , in the film and to the film thickness, δ , as:

$$J = \frac{D}{\delta} \Delta C \tag{2.3}$$

where *D* is the diffusivity of the transferring species and D/δ is the mass transfer coefficient, *k*. At steady state, the flux across the gas and liquid films are equivalent and equation 2.3 can be re-written for each film:

$$J = k_G (C_G - C_{Gi}) = k_L (C_{Li} - C_L)$$
(2.4)

where k_G and k_L are the gas and liquid film mass transfer coefficients, respectively. The overall mass flux from the gas to the liquid phase is:

$$J = K_L(C^* - C_L)$$
 (2.5)

where C^* is the saturation concentration of the diffusing species in the liquid in equilibrium with the gas phase and K_L is the overall mass transfer coefficient in the liquid. Further simplification leads to the final form:

$$\frac{dC}{dt} = k_L a(C^* - C) \tag{2.6}$$

where C is the dissolved gas concentration in the liquid at time t and a is the gas-liquid interfacial area per unit volume of the liquid.

Bubble size becomes important when considering the gas-liquid mass transfer issues in chemical processes. In the kraft pulping process, the gas-liquid mass transfer is important in a number of unit operations, including the oxygen delignification and extractive oxidation stages. Due to the low solubility of oxygen in water, it must be applied to pulp suspensions directly in gas phase. When the gas is introduced into a pulp suspension, the gas must first be well mixed with the pulp suspension. The gas then diffuses from the bulk gas phase to the gas-liquid interface then transfer from the gas phase to the liquid phase, diffuse from the interface to the bulk liquid phase, diffuse from the bulk liquid to a fibre surface, diffuse through the fibre pores and matrix to a reaction site of the pulp fibre where finally it reacts with the lignin and other non-cellulosic materials. These steps occur in series and the slowest step will ultimately control the overall mass transfer performance. Figure 2.10 schematically represents the transfer of oxygen in the gas-liquid-fibre reaction.



Figure 2.10: Mass transfer in oxygen delignification (Iribarne and Schroeder, 1997)

Upon reaction, the lignin product from the fibre dissolves to the bulk liquid phase. The overall rate of mass transfer per unit volume is governed by the volumetric gas-liquid mass transfer coefficient, $k_L a$, assuming that the gas side resistance is negligible. The factors affecting $k_L a$ is mostly due to the interfacial area parameter, a. In the oxygen delignification case, the surface area of an oxygen bubble in the gas-pulp suspension mixture has an effect on the effectiveness of the delignification. In a well mixed gas-pulp suspension, oxygen gas is uniformly dispersed in the pulp suspension resulting in a large gas-liquid interfacial surface area, a, and ensures uniform gas-pulp suspension dispersion and an overall fast and efficient reaction in the system. On the other hand, in a poorly mixed gas-pulp suspension, the non-uniform dispersion may lead to reduced interfacial surface area and result in a non-uniform, slow and inefficient chemical reaction with the reaction site in the fibre.

The separation of the volumetric mass transfer coefficient, $k_L a$, into separate parameters k_L and a should be taken under consideration for a better understanding of the

gas-liquid mass transfer mechanism in order to identify which parameter (k_L or a) ultimately controls the mass transfer rate.

Many correlations exist in literature for predicting the mass transfer coefficient in bubble column reactors; however, it is important to note that they are only applicable for certain conditions. The majority of the correlations deal with air-water systems in the absence of solid particles. However, most industrial applications typically have a high solids concentration in order to achieve high reaction rates (Kantarci *et al.*, 2005).

Factors that will affect the efficient mass transfer include: superficial gas velocity, liquid phase properties, solid concentration, bubble properties, column dimensions, gas sparger, and operating conditions (such as temperature and pressure).

Studies of the mass transfer in pulp suspensions are scarce. Bennington *et al.* (1997) measured mass transfer rates in a high-shear laboratory mixer under mediumconsistency bleaching conditions and results showed that the volumetric mass transfer coefficient, $k_L a$, varied with power dissipation and fibre mass concentration. The results in pulp suspensions were an order of magnitude lower compare to water under the same operating conditions and decreased further with increasing solids concentration. Rewatkar and Bennington (2000) conducted further studies in the high-shear mixer for low and medium consistency pulp suspensions with visual observations using high-speed videos and still photographs.

The effect of oxygen mass transfer on the delignification in an industrial retention tower was simulated in Van Heiningen *et al.* (2003). As a result of the study, the simulation suggested that industrial oxygen mixers are not effective in distributing oxygen and that the delignification predominantly occurred in the tower, as opposed to the mixer. The results showed that the mass transfer increased significantly when the consistency increased from 8 to 14%. The limitations of the mass transfer in the tower, however, could not be clearly identified.

Rewatkar and Bennington (2004) studied the volumetric mass transfer rate for pulp suspensions, this time in a pulp retention tower under batch operating conditions. Results showed that at low mass concentrations (C_m less than 2 or 3%) the tower portrayed bubble column behaviour and the mass transfer was comparable to that measured for bubble columns but decreased as the suspension concentration increased.

The mass transfer reached a minimum for consistencies between 3 and 6% but as the suspension concentration increased beyond 6%, $k_L a$ increased as well.

2.6 Electrical Resistance Tomography (ERT)

The term *tomography* originates from the Greek words, *tomos* meaning slice or cross-section and *graph* meaning image or picture. Electrical resistance tomography (ERT) is a method that employs electrical measurements in order to obtain information of the contents of process vessels and pipelines. A two-dimensional cross-sectional image of the region of interest is produced in real time.

Tomographic methods can be dated back to the 1920's. Geophysicists made use of resistivity imaging in order to better describe oil-bearing rock in the earth. The method employed inserting arrays of metallic electrodes into the ground, applying a current between adjacent electrodes, and then measuring the resultant voltage response of the other electrodes. A map of the sub-surface layer could then be produced. This method is still used today with the implementation of cross bore hole tomography in which vertical poles, containing annular electrodes are inserted into bore holes several hundred metres apart around the region of interest. Not until the 1970's has the method of tomographic resistivity measurements been employed in the medical field as an alternative to the x-ray scanner.

Electrical resistance tomography has more recently become increasingly popular in the chemical and process industries since valuable information of the process can be gathered by this non-invasive technique as opposed to other measurement sensors which may disturb the flow pattern of the system and therefore not accurately represent the process. Information that may be determined using this technique include the density, concentration, temperature, velocity, and local void fraction in the measurement plane.

There are two main advantages to this method. First, tomographic methods do not invasively disturb the natural operation of the system, and second, the region of interest is captured at many angles in the measurement plane so that an accurate representation of the cross-sectional region of interest can be achieved. However, in order to capture an accurate image, tomographic methods must have high spatial and temporal resolution, issues which have been addressed as limitations described in literature (Toye *et al.*, 2005; Stephenson *et al.*, 2007; Dickin and Wang, 1996; Daily *et al.*, 2004). Although it does offer unique capabilities other imaging techniques cannot. Some other advantages include:

- Easy automation
- Fast data processing capabilities
- Reasonable cost.

In flow applications for example, it can be used to characterize flow regimes, measure the concentration of different components in the multiphase flow stream, as well as detect foreign objects present in the measurement plane.

A series of electrodes are arranged around the inside edge of a vessel in such a way that it makes electrical contact with the contents of the vessel but does not disturb the flow of material inside the vessel. The electrodes, which are made of a material largely dependent on the process application but more conductive than the fluid being imaged, are connected to the data acquisition system via co-axial cable which reduces interference and noise of the surrounding environment. Figure 2.11 shows the arrangement of electrodes around a process vessel at equal intervals.



Figure 2.11: Schematic diagram of electrode arrangement around a process vessel (ITS, 2003)

The data acquisition system shown in Figure 2.12 (ITS P2000, Manchester, UK) injects a current between a pair of electrodes and measures the resultant voltage difference between the remaining electrode pairs according to a pre-defined measurement protocol (ITS, 2003).



Figure 2.12: Picture of ITS P2000 tomography unit

Tomographic measurement technique is accomplished by two consecutive steps. First, integral measurements of the cross-sectional measurement plane (region of interest) are made by the use of the tomographic sensor coupled with the data acquisition system, then, the reconstruction of the integral measurement values are performed to produce an image of the object in the measurement plane (region of interest). Figure 2.13 shows the schematic of a tomographic system which is composed of three main parts: sensors, data acquisition system, and image reconstruction system (computer). Figure 2.14(a) shows the steps performed to obtain a reconstructed image and 2.14(b) shows the tool used for the respective measurement.



Figure 2.13: Schematic of an ERT system (Dickin and Wang, 1996)



Figure 2.14: Schematic description of tomography measurement steps (Petritsch et al., 2005)

Following the data acquisition of the object boundary, appropriate image reconstruction is performed to develop the cross-sectional distribution of the electrical conductivity within the measurement plane. The plane is divided into a square 20 x 20 grid of pixels representing the interior cross-section of the vessel, for which some pixels lie on the outside of the vessel circumference therefore the reconstructed image is obtained from 316 of the 400 pixels as shown in Figure 2.15. Each pixel contains

information on the conductivity at that location according to a pre-defined measurement protocol.



Figure 2.15: ERT image reconstruction grid (ITS, 2003)

ERT has become an increasingly powerful tool in various industrial applications for visualization of the characteristics of the fluid dynamics in multiphase systems. It has been applied as an extension to more conventional measurement techniques. For example, ERT has been used to analyze bubble behaviours such as the bubble rise velocity, bubble size, and bubble shape, (Wu *et al.*, 2005; Jin *et al.*, 2007), as well as a means to identify the hydrodynamics of bubble columns in determining the flow regime and gas holdup (Fransolet *et al.*, 2001; Fransolet *et al.*, 2005; Dong *et al.*, 2005). In addition to determining these parameters, ERT has also provided a means of visually validating these characteristics.

CHAPTER 3

BUBBLE CHARACTERISTICS IN A RECTANGULAR VESSEL

Many industrial reactors are cylindrical in shape. The rationale for a cylindrical configuration is to provide for mechanical stability due in cases where high operating pressures are present as well as to create symmetry for the uniform distribution of reacting species. This becomes especially important for bubble columns where it is critical to have a uniform distribution of gas bubbles throughout the reactor volume. However, when studying the bubble rise characteristics, it is often difficult to obtain an accurate visual representation of the bubble due to the geometry of the vessel and the characteristic nature of its contents. In the pulp and paper industry, the presence of opaque fibres makes it difficult to observe bubble characteristics; however, bubble size is an important parameter in understanding the mass transfer ability. For example, during the bleaching stage in the kraft pulping process, it is desirable to have small, uniformly sized, homogeneously distributed gaseous phase (such as oxygen) throughout the system to maximize the bubble surface area for greatest mass transfer effect. Since this reaction occurs in a cylindrical vessel having a circular cross-section, it is difficult to measure or even observe the shape of the bubbles rising through an opaque fibre suspension. For this reason, a rectangular vessel (two-dimensional vessel) is used to observe the behaviour of the contents of the vessel and is chosen for its accessibility for optical observations. The shape, size, and rise velocity of air bubbles is determined in a multiphase system and can be used to predict the behaviour of the gas-liquid-fibre suspension in a cylindrical vessel.

3.1 Literature Review

Rectangular vessels have been widely used in many studies to describe the hydrodynamic behaviour in multi-phase systems (Krepper *et al.*, 2007; Zaruba *et al.*, 2005(a,b); Buwa and Ranade, 2005; Buwa and Ranade, 2004; Vandu *et al.*, 2004; Magaud *et al.*, 2001; Krishna and van Baten, 1999). However, few studies report the bubble characteristics in systems with pulp suspensions as the continuous phase. Reese

et al. (1996) described bubble characteristics in water and in pulp suspensions at low mass concentraitons (ranging from 0.1 to 1.0%) in both rectangular and cylindrical vessels. An intrusive light transmittance probe was used to measure the bubble passing frequency and period in a cylindrical column to determine the presence of bubbles in a specified zone. A non-intrusive particle image velocimetry (PIV) technique was used to determine the bubble size and bubble rise velocity, as well as to qualitatively observe the bubble shape in a rectangular column. As a result of their study, they concluded that even at low pulp mass concentrations, the behaviour of the bubble deviates from the behaviour of that in water and the deviation is more significant as the pulp mass concentration increases.

Heindel and Garner (1999) examined bubble size distributions in northern bleached softwood kraft (NBSK) pulp suspensions ranging from 0 (water) to 1.5% in a 1 m high bubble column and 20 cm by 2 cm rectangular cross-section by applying flash xray radiography (FXR). X-ray radiography is a technique that applies short intense bursts of x-rays to capture dynamic events on film. With as little as 0.5% NBSK, a flow pattern change resulted with the development of larger bubbles which promoted churn-turbulent flow conditions. They concluded that as the NBSK consistency increased, differences in the cumulative number density of the bubble size distribution increased. The bubble size distribution was described by a single lognormal distribution when the bubble diameters were less than 12 mm.

Flash x-ray radiography was applied again by Garner and Heindel (2000) to determine the effect that different fibre types had on bubble size. Three different pulp suspensions with varying fibre lengths but at a constant consistency of 1% were studied in the identical experimental apparatus as Heindel and Garner (1999). Results showed that the general bubble population (number of bubbles in the vessel at any given time) decreased with increasing fibre length, the bubble population with diameters greater than 12 mm increased with increasing fibre length, and the bubble size increased with increased with increasing fibre length.

Lindsay, Ghiaasiaan, and Abdel-Khalik (1995) described the macroscopic properties and flow regimes of gas-liquid-fibre suspensions in batch and co-current configurations in two cylindrical columns. With the batch configuration a transition from

bubbly flow to churn flow was observed with the presence of pulp fibres and an increase in bubble coalescence was noted. With the co-current flow, bubbly, plug, and transitional flow regimes were identified; however, they could only be observed near the wall of the vessel. Further discussion is provided in Chapter 4 regarding cylindrical vessels.

Describing bubble behaviour in pulp fibre suspensions at low mass concentrations can be applied to understand the hydrodynamics in flotation deinking operations, a separation process to remove ink from fibres for the production of recycled paper. Most other industrial pulping operations operate at much higher pulp mass concentrations but little is known about their behaviour.

This study provides preliminary insight into the behaviour and characteristics of bubbles rising in a stationary liquid phase (water) or pulp suspension (wood fibres and water mixture) of pulp mass concentrations up to 7%. It can be used to describe the three-phase behaviour in oxygen delignification towers present in the kraft pulping process.

3.2 Experimental Apparatus

Experiments were conducted in a two-dimensional Plexiglas bubble column of rectangular cross-section with sparged air as the gas phase and water, as the stationary liquid phase, or pulp suspensions of varying mass concentrations and pulp types as the stationary suspension phase. The rectangular vessel is used to study the hydrodynamics of bubbles in pulp suspensions by examining the bubble shape, size, behaviour, and rise velocity as a function of pulp type, pulp consistency and gas flow rate.

The experimental apparatus (Figure 3.1) consists of a rectangular channel made of two 1.1 cm thick by 56.5 cm long transparent Plexiglas sheets and two 2.2 cm thick by 2.5 cm wide rectangular transparent Plexiglas sections to complete the two-dimensional vessel. The dimensions of the internal volume of the vessel are 90 cm in height, 56.5 cm in length, and 2.5 cm in width. A Swagelock fitting located at the bottom centre of the channel allows for filtered compressed air to flow through the vessel via 1/4-inch diameter flexible polyethylene tubing and into a 0.3 cm diameter (0.1 cm inside diameter) stainless steel tubing protruding midway into the gap width in the vessel for air introduction. The presence of a single portal for air introduction was used in order to observe individual bubbles in the channel and to limit bubble-bubble interaction. The air flow rate is measured using a mass flow meter (Cole-Parmer Model 32915-68, Vernon Hills, IL). The vessel is also equipped with a 1.75 cm diameter pipe connected to a valve for draining the contents of the vessel when it is filled with water or low consistency pulp suspensions ($C_m \leq 1\%$); however, the channel must be inverted in order to drain higher consistency pulp suspensions due to pipe plugging issues.



Figure 3.1: Schematic of rectangular vessel for 2-D experiments

The channel was backlit with a diffuse light source and each test was recorded using a NAC HSV-1000 high-speed video system (NAC, Simi Valley, CA) at 500 frames

per second and 400+ lines horizontal resolution. Video data was then analyzed to determine bubble shape, size, and rise velocity.

3.3 Experimental Methods and Techniques

Water and bleached elemental chlorine free softwood and hardwood kraft pulps supplied by Domtar Inc. (Windsor, QC) were used for all tests. The bleached softwood (SW) pulp was made of a northern bleached softwood kraft (NBSK) pulp containing a high proportion of Jack Pine had a length-weighted fibre length of 2.34 mm, a coarseness of 0.16 mg/m with the yield stress described by τ_y (Pa) = $12.8C_m^{2.03}$. The bleached hardwood (HW) pulp was predominantly composed of Maple wood fibres with fibre length of 0.70 mm, a coarseness of 0.079 mg/m with the yield stress described by τ_y (Pa) = $1.23C_m^{3.00}$. The average fibre length and fibre coarseness for both pulps were determined in-house using the Fibre Quality Analyzer (FQA) (OpTest Equipment Inc., Hawkesbury, ON). Other typical pulp properties as reported by the supplier are presented in Table 3.1.

Pulp	Fibre	Fibre	Brightness	Dirt	Viscosity	DCM	Ash	Air
Туре	Length	Coarseness	(ISO)	(ppm)	(cP)	Extractives	Content	Dry
	(mm)	(mg/m)				(%)	(%)	(%)
SW	2.34	0.160	89	< 5	16-22	0.021	0.30	86
HW	0.70	0.079	90	0.1	7-9	0.015	0.24	93

The pulp suspensions were examined at mass concentrations of 0 (water), 1, 2, 4, and 7%. The channel was filled with 9.5 litres of the sample, giving a height of 67.3 centimetres and air was introduced to the vessel at flow rates ranging 0 - 20 litres per minute. The channel was backlit with an intense photographic-quality lamp with a sheet of PVC plastic having diffusive properties placed between the channel and the lamp in order to reduce the glare produced by the light so that a clear video image could be obtained for subsequent analysis. Each trial was recorded using a NAC High Speed

Video System HSV – 1000 (NAC, Simi Valley, CA). Video data was then analyzed to determine the bubble behaviour, shape, size, and rise velocity. The experimental procedure adopted was as follows:

- 1. Prepare pulp suspensions having mass concentrations of 1, 2, 4, and 7% giving a total volume of 9.5 litres.
- 2. Fill the channel with the desired pulp suspension.
- 3. Mix the contents of the vessel ensuring that a uniform suspension is achieved and that no air bubbles are present in the suspension.
- 4. Adjust the lamp, PVC diffusive sheet, and high speed video camera for best image-capture.
- 5. Turn on the air valve and set to the desired flow rate.
- Record for approximately 10 seconds using the high speed video camera at 500 frames per second.
- 7. Repeat step 3.
- 8. Change the air flow rate and repeat steps 6 and 7 for all desired air flow rates.
- 9. Empty the contents of the channel and clean thoroughly with water.
- 10. Repeat steps 2-9 for all pulp consistencies and air flow rates.

3.4 Data Analysis

3.4.1 Velocity

Using the playback feature of the HSV – 1000 video system, bubbles can be isolated and followed up the screen as a function of time in order to determine the bubble behaviour, rise velocity, bubble shape and bubble size. The bubble rise velocity (U_b) was determined by isolating a single bubble on the monitor of the HSV – 1000 video system, recording the distance travelled from height H_1 to H_2 and the time required to travel the distance. The bubble rise velocity (U_b) was calculated as follows:

$$U_{b} = \frac{H_{2} - H_{1}}{t_{2} - t_{1}} \tag{3.1}$$

where H_1 is the bubble height at position "1", H_2 is the bubble height at position "2", t_1 is the time at position "1", t_2 is the time at position "2", and U_b is the bubble rise velocity.

3.4.2 Size

The bubble size in the rectangular channel was determined by measuring with a ruler the two-dimensional bubble length and height as shown in Figure 3.2.



Figure 3.2: Bubble dimension description in rectangular channel

Bubble size was determined once the bubble had sufficient time to develop following the air port introduction. Therefore, the measurements of the bubble was often taken to be at the centre or further up the channel, and not at the location near the air port introduction since the bubble shape and size near this location does not accurately represent the bubble characteristics when the bubble has fully developed.

3.4.3 Shape

The playback feature of the HSV – 1000 video system was also used to determine the bubble shape. The shape regime map (Figure 3.3) for bubbles in motion through liquids (Clift *et al.*, 1978) was used to characterize the shape of the bubbles in this study.



Figure 3.3: Description of bubble shape in motion in liquids (Clift et al., 1978)

The graphical correlation shown in Figure 3.3 is used to predict the bubble shape in terms of the Eotvos number, Eo, Morton number, M, and the Reynolds number, Re.

The observed results can then be compared to the predicted results calculated by using the dimensionless numbers and then determining the regime the value falls within. The dimensionless terms are defined as:

$$Eo = \frac{g \,\Delta \rho \,d_e^2}{\sigma} \tag{3.2}$$

$$M = \frac{g\,\mu^4 \Delta \rho}{\rho^2 \sigma^3} \tag{3.3}$$

$$Re = \frac{\rho d_e U}{\mu} \tag{3.4}$$

Another use for the correlation is to estimate the terminal velocity of the bubble in the suspension. The Eotvos and Morton numbers are determined and located on the graph, and then at the point of intersection, the Reynolds number is read off the y-axis. The velocity is then determined from equation 3.4 using the Reynolds number from the graph and the known values for the density, ρ , diameter of volume equivalent sphere, d_e , and viscosity, μ . This could only be determined for bubbles in water.

Pulp suspension rheology is a very complex subject and because of this, the viscosity and the surface tension of pulp suspensions are difficult to determine experimentally and not widely reported in the literature. Normal viscometers are not able to provide sufficient mixing required for maintaining a homogeneous uniform suspension of water and fibres, leading to erroneous viscosity results which are more representative of the liquid phase in the suspension rather than the suspension as a whole (Chase *et al.*, 1989).

3.5 RESULTS AND DISCUSSION

The introduction of a gas phase into a liquid phase leads to the formation of a bubble. The natural tendency of a gas bubble introduced to a liquid medium is to rise towards the surface. As the gas bubble rises, it carries the liquid along with it and forces the liquid to reverse flow direction near the walls of the vessel thus forming recirculation cells. This phenomena is described in Figure 3.4.



Figure 3.4: Flow behaviour in a bubble column: (left) uniform bubble rising behaviour and liquid recirculation, (right) oscillating bubble rising behaviour and non-uniform liquid recirculation (Diaz *et al.*, 2006).

The shape, size, behaviour, and rise velocity of the bubble will depend upon the properties of both the gas and the liquid mediums. Solid material, such as pulp fibres, dispersed in the liquid phase will also affect the bubble characteristics, in addition, the behaviour of the bubble in different mass concentrations of pulp suspensions as well as pulp types will also be factors affecting the bubble characteristics, as will be described subsequently.

3.5.1 Bubble Characteristics in Water

Bubbles were observed in water with air flow rates ranging from 1.50 to 10.0 litres per minute. Bubble rise velocities increased as the gas flow rate increased with rising velocities in the range 0.58 to 0.99 m/s. The characteristic diameter chosen as the bubble width (as in Figure 3.2) ranged from 2.0 to 4.1 centimetres in water. At low gas flow rates (Q < 3 L/min), the air bubbles are consistently spherical in shape with minimal

bubble breaking or coalescing as they rise in the liquid phase up the channel. As the air flow rate is increased, the bubble shape starts to deform and grow into larger slugs of bubbles rising up the channel with some being spherical-capped bubbles and the presence of bubble breaking and coalescing increases as the air flow rate increases. It was also observed that recirculation of the liquid phase along with the presence of small bubbles was more prevalent as the air flow rate increased. Figure 3.5 shows two cases of bubbles rising in water at slow and fast air flow rates.



Figure 3.5: Bubbles in water (a) air flow rate = 1.6 L/min, (b) air flow rate = 9.9 L/min

Zaruba *et al.* (2005 (a,b)) found that with a superficial gas velocity of 1 mm/s (corresponding to 0.12 L/min in a rectangular bubble column of 10 cm by 2 cm cross section and 1.5 m height), the bubble-bubble interactions were uncommon such that the dispersed bubbly flow regime was observed and the bubble axial velocity was in the range of 0.20 to 0.26 m/s.

3.5.2 Bubble Characteristics in Softwood Kraft Pulp Suspensions

Softwood pulp suspensions were investigated at pulp mass concentrations of 1%, 2%, and 4%. The air flow rates examined ranged from 1.50 to 10.0 litres per minute.

The bubble rise velocity in a $C_m = 1\%$ softwood suspension ranged from 0.24 to 0.66 m/s. The bubble shape at low gas flow rates (Q < 3 L/min) resembles those in water, spherical in shape with some wobbling bubbles. At higher gas flow rates, the bubble shape progresses into elliptical shaped bubbles and a few dimpled ellipsoidal-capped bubbles. Uniform liquid recirculation is observed as the bubble rises in the suspension.



Figure 3.6: Bubbles in 1% softwood pulp (a) air flow rate = 1.5 L/min, (b) air flow rate = 10.0 L/min

The bubble rise velocity in a $C_m = 2\%$ softwood suspension ranged from 0.43 to 0.67 m/s. The bubble shape at low gas flow rates begins as deformed spherical bubbles but as time increases and the gas rises through the suspension, a trail of void space corresponding to a mixture of gas and water phases is left behind and becomes the path of the subsequent bubble to follow. This suspension becomes non-uniform due to the phase

separation and regions of lower pulp mass concentration are formed. After a short period of time (t < 2 s) following phase segregation, no specific bubble shape can be observed as there is only a region of void space in the centre of the suspension with no liquid recirculation. The bulk of the suspension then becomes resistant to the motion of the air flowing through it at that particular air flow rate. At higher gas flow rates; however, the bubble shape is large, spherical, and more deformed and it oscillates as it rises with non-uniform liquid recirculation (see Figure 3.4).



Figure 3.7: Bubbles in 2% softwood pulp (a) air flow rate = 1.4 L/min, (b) air flow rate = 9.9 L/min

In a $C_m = 4\%$ softwood suspensions, distinct bubble shape was no longer observed and the bubble rise velocities could not be measured. Void space was observed at the centre of the channel providing a pre-determined path for the air to flow up through the suspension at all gas flow rates with no presence of liquid recirculation.



(a) (b) Figure 3.8: Bubbles in 4% softwood pulp (a) air flow rate = 1.5 L/min, (b) air flow rate = 10.0 L/min

The diameter (width as in Figure 3.2) of the gas bubbles in softwood pulp ranged from 1.9 to 5.1 centimetres (compared to 1.9 to 4.1 cm in water). The larger size of the bubbles in softwood suspensions confirms that in the presence of fibres, bubble coalescence increases.

In a previous study with the identical rectangular experimental set-up, Pineault and Bennington (2002) found the bubble rise velocity in stone groundwood (SGW) with pulp mass concentrations, $C_m \le 4\%$ to be in the range of 0.4 to 0.8 m/s. The bubble size increased with suspension mass concentration and gas flow rate and the bubble diameters that resulted were between 1 and 5 centimetres.

3.5.3 Bubble Characteristics in Hardwood Kraft Pulp Suspensions

Hardwood pulp suspensions were investigated at mass concentrations of 1%, 2%, 4%, and 7% in water. The air flow rates examined ranged from 1.20 to 20.0 litres per minute.
The bubble rise velocity in a $C_m = 1\%$ hardwood suspensions ranged from 0.55 to 1.0 m/s. Bubbles were mostly spherical in shape with few spherical-capped bubbles at low gas flow rates and progressed to ellipsoidal and spherical- or dimpled ellipsoidal-capped bubbles at higher gas flow rates with some wobbling as well. Uniform bubble rising behaviour and uniform liquid recirculation was observed at all air flow rates.





Bubble rise velocity in a $C_m = 2\%$ hardwood suspensions ranged from 0.33 to 0.71 m/s. Air bubbles were spherical and ellipsoidal at times at low gas flow rates with minimal uniform gas phase rising behaviour and uniform liquid recirculation. At higher gas flow rates, the bubble size increased and spherical-capped and dimpled ellipsoidal-capped bubbles were observed with increased uniform liquid recirculation as a result of the higher air flow rates. Bubble breaking and coalescing was also more prevalent at higher gas flow rates.



Figure 3.10: Bubbles in 2% hardwood pulp (a) air flow rate = 1.3 L/min, (b) air flow rate = 9.3 L/min and, (c) air flow rate = 20.0 L/min.

Bubble rise velocity in a $C_m = 4\%$ hardwood suspensions ranged from 0.47 to 0.66 m/s. Deformed spherical bubbles at low gas flow rates were observed and at higher gas flow rates, the bubble shape is large and spherical and more established (rises without deforming). Bubble coalescence is more prevalent than bubble breaking at high gas flow rates.



Figure 3.11. Bubbles in 4% hardwood pulp (a) air flow rate = 1.2 L/min, (b) air flow rate = 9.1 L/min and, (c) air flow rate = 19.1 L/min.

No distinct bubble shape was observed in 7% hardwood suspensions and the bubble rise velocities could not be measured. Void space was observed at the centre of the channel providing a path for the air to flow up through the suspension at all gas flow rates with no presence of liquid recirculation. The diameter (width as in Figure 3.2) of the gas bubbles in hardwood pulp with mass concentrations between 1 and 4% ranged from 1.7 to 7.1 centimetres.

Overall bubble size results for water, softwood and hardwood pulps ranged between 1.9 to 4.3 cm, 1.5 to 6.3 cm, and 1.4 to 7.1 cm, respectively for gas flow rates between 1.50 to 10.0 L/min.

Please refer to Appendix A1 for the complete set of still images of all trials.

3.5.4 Summary of Results

Bubble characteristics were observed in water and in softwood and hardwood pulp suspensions with varying mass concentrations. Results for water were comparable to that in literature and were different compared to pulp suspensions at the same gas flow In addition, a significant difference was observed between the softwood and rate. hardwood pulps. During the preparation of the hardwood pulp at the same consistency as the softwood pulp, it was observed that the suspension was more uniform and homogeneous, even at higher mass concentrations, compared to that of the softwood pulp suspension. With the same method of preparation, the softwood pulp suspension formed flocs of fibres in the suspension (regions of softwood fibres swelled with water and regions with water alone), rendering the suspension less uniform. This is likely due to the fact that the length of softwood fibres are longer (2.34 mm for softwood fibres compared to 0.70 mm for hardwood fibres, see Table 3.1) and as a result, the fibres are more easily entangled with each other forming bundles of fibres in a suspension, and thus the bubble characteristics behaved accordingly. Reese et al. (1996) found that even at very low pulp mass concentrations ($C_m = 0.1\%$) the hydrodynamic behaviour of the suspension behaves differently from gas-liquid systems. The bubbles were significantly affected by the presence of pulp fibres and were flatter and rose more slowly in the pulp slurry than in water. In addition, Heindel and Garner (1999) also found that the flow pattern behaviour of bubbles changed from bubbly to churn-turbulent with as little as 0.5% NBSK and the bubble population decreased as the NBSK consistency increased. These findings were consistent with those found in this study. Bubbles were significantly affected by the presence of pulp fibres and the effect increased as the pulp mass concentration increased. In addition, this study showed that when different fibre types were used (softwood or hardwood) the bubble size and shape were affected even at the same pulp mass concentrations. This was also observed and described in Garner and Heindel (2000).

Figure 3.12 shows a graphical representation of the bubble rise velocity as a function of gas flow rate. In all cases, the bubble rise velocity increases as the gas flow rate increases. The bubble rise velocity in water is, for the most part, consistently higher

than all other cases since the upward motion of the bubble is not impeded by any solid material present (pulp fibres).

An interesting observation occurred with $C_m = 2\%$ compared to $C_m = 1\%$ softwood. The bubble rise velocity of the 2% suspension was higher than the 1% up to gas flow rates of 6 L/min. In the 1% trial, a trail of void space was left behind the previous bubble making the subsequent bubble rise faster following the same path which is of less resistance. As time progresses, the bubble is no longer of distinct shape, just a void space in the suspension providing a region for the air to flow up through the suspension. This can be attributed to the fact that the air flow rate is low such that it cannot move past the pulp fibre flocs. It was confirmed that the pulp suspension was stationary at low air flow rates, but when the air flow rate was increased, the bubble rise velocity for 1% suspension was then higher than that of the 2% suspension. In this case, there was motion of the liquid phase, proving that the air flow rate was sufficient enough to pass by the fibre network which had otherwise impeded the flow at the lower air flow rates.



Figure 3.12: Bubble rise velocity versus air flow rate in water and pulp suspensions

Table 3.2 and 3.3 presents the bubble rise velocity and bubble size data collected for water, softwood kraft pulp, and hardwood kraft pulp. As expected, the bubble size consistently increases as the gas flow rate increases for all liquid mediums. The bubble size data shows that the size of the bubbles is greater in fibre suspensions with bubble sizes in hardwood suspensions consistently greater than bubble sizes in softwood suspensions. This can be attributed to the fact that the fibres of hardwood are shorter and smaller than those of softwood and thus the bubbles can manoeuvre past the fibres more easily rather than being entangled in the network of fibres that are present in softwood suspensions, especially at higher consistencies.

	Bubble Rise Velocity (m/s)						
Air Flow (L/min)	Water	$C_m = 1\%$		$C_m = 2\%$		$C_m = 4\%$	
		SW	HW	SW	HW	HW	
1.5	0.58	0.24	0.55	0.43	0.33	0.47	
3.0	0.76	0.35	0.64	0.47	0.42	0.50	
4.5	0.79	0.42	0.72	0.51	0.48	0.54	
6.0	0.77	0.54	0.81	0.56	0.54	0.57	
8.0	0.82	0.63	0.92	0.62	0.63	0.61	
10	0.99	0.66	1.0	0.67	0.71	0.66	

Table 3.2: Summary of bubble rise velocity results in water and pulp suspensions

Table 3.3: Bubble size and shape in water and pulp suspensions at various air flow rates

Air Flow (L/min)	Bubble Size (w,h)*(cm) and Shape°						
	Water	$C_m = 1\%$		$C_m = 2\%$		$C_m = 4\%$	
		SW	HW	SW	HW	HW	
1.5	2.0, 1.9 ^s	1.9, 1.9 ^s	1.7, 1.4 ^s	2.3, 3.1 ^w	2.7, 2.4 ^e	2.5, 2.8 ^s	
3.0	2.9, 3.2 ^s	2.5, 2.3 ^s	$2.3, 2.2^{e,sc}$	2.7, 3.4 ^w	$3.1, 2.6^{e.sc}$	$3.3, 4.0^{s,w}$	
4.5	2.9, 3.2 ^s	2.6, 2.4 ^s	$2.9, 2.0^{e,sc}$	$1.5, 2.7^{w}$	$3.0, 2.3^{e,sc}$	$3.2, 5.0^{\text{s.w}}$	
6.0	3.8, 2.9 ^e	3.7, 2.8 ^{e,w}	3.1, 2.8 ^{e,sc}	3.1, 4.0 ^w	4.2, 2.8 ^{de}	$5.8, 6.0^{s,w}$	
8.0	$4.0, 4.0^{\rm s}$	$4.1, 3.3^{e,w}$	4.1, 3.8 ^{de}	3.6, 5.2 ^w	$5.8, 3.0^{de}$	$6.6, 6.8^{s,w}$	
10	4.1, 4.3 ^s	$4.5, 3.6^{e,w}$	4.9, 4.1 ^{de}	5.1, 6.3 ^w	7.1, 3.4 ^{de}	5.7, 6.9 ^{s,w}	

*w and h are the horizontal and vertical bubble sizes, respectively, with ± 0.1 cm margin of error.

^obubble shape designations (Clift *et al.*, 1978): de - dimpled ellipsoidal capped; e – ellipsoidal; s - spherical; sc - spherical-capped; w – wobbling.

3.5.5 Comparison With Graphical Correlation

The Eotvos number, Eo, the Morton number, M, and the Reynolds number, Re, were determined for all water trials. The Eotvos number and the log of the Morton number were used to determine the predicted shape regime that the bubble belonged to according to the graphical correlation (Figure 3.3). According to the graphical correlation, the majority of the bubbles should lie in the spherical-capped regime with few approaching the wobbling, ellipsoidal, and skirted bubble regime. These predictions are fairly accurate, as the boundaries of the shape regimes in the graphical correlations are somewhat arbitrary as stated in Clift *et al.* (1978).

In addition to estimating the shape regime using the graphical correlation, it may also be used to estimate the terminal velocity; however, as stated by Clift *et al.* (1978), more accurate correlations are usually available. The results presented in Table A2.3 in Appendix A2 shows significant discrepancies between the measured and predicted bubble rise velocity data for water with a percent error as high as 48%. More accurate predictive correlations for the terminal velocity should be used if one wanted to estimate the bubble terminal velocity.

CHAPTER 4

GAS HOLDUP AND MASS TRANSFER IN A CYLINDRICAL VESSEL

The cylindrical vessel, which is the main focus of this study, is used to study the hydrodynamics and mass transfer effects of air as the gas phase and pulp suspensions of different types and consistencies as the liquid-solid phase operated in batch mode (referred to as the bubble column). The purpose of this study is to understand the behaviour of the suspension in pulp retention towers and help explain the limitations that exist in some industrial towers (discussed in Chapter 2).

4.1 Literature Review

Bubble columns are unique type of reactors in that they operate without mechanical agitation. In bubble columns, gas is dispersed by nozzles or spargers which provide for the mixing requirements in the tower. For effective operation, re-dispersion of the gas by static mixers such as perforated plates along the tower is essential (Perry and Green, 1997). Bubble columns are preferred over other types of reactors because of their simplicity of operation, low maintenance and operating costs due to the lack of moving parts, and good heat and mass transfer. They are often used when slow reactions are involved and when the mass transport resistance is located on the liquid side (Wild et al., 2003), in other words, when the rate of reaction is smaller than the rate of diffusion. The advantages of bubble columns is the absence of moving parts, the ability to handle solid particles without erosion or plugging, good heat transfer at the wall or coils, high interfacial area, and high mass transfer coefficients. The disadvantages include the backmixing of the liquid phase and at times the gas phase as well, which may result in poor selectivity for complex reactions. Another drawback is the high pressure drop due to the static head of the liquid. Also, if the height-to-diameter ratio is large (>15), the effective interfacial area decreases rapidly (Perry and Green, 1997).

In bubble column reactors, gas bubbles flow upward through a slower moving liquid. The rising bubbles draw liquid in their wakes and thereby induce back-mixing in

the liquid with which they have come in contact. The flow of the gas and the liquid in the column differ depending on the gas and liquid velocities, gas distributor type, column diameter, and physical properties of the liquid. Since it is not possible to describe the local flow of both the gas and liquid phases in the tower, a global description of the flow as flow regimes is most often described in literature (Xie et al., 2003; Kantarci et al., 2005; Zehner and Kraume, 2005; Garner and Kerekes, 1980). Bubble behaviour has a direct bearing on the hydrodynamics, mass transfer, and overall reactor performance. The description and design of multiphase reactors still to a great extent rely on empirical methods and correlations based on conditions that most closely simulate industrial practice. Although extensive research exists on bubble columns with two and three phase systems, there is limited research in systems containing pulp suspensions, most likely due to its complex and unique rheology. Consequently, reliable measuring techniques are needed to accurately describe and examine multiphase reactors. One classification method of classifying measurement techniques is: global or local measurements. Global measurements describe a parameter depicting the overall system, whereas local measurements describe a parameter to a specific region in the system.

One of the parameters in examining reactor performance is to measure the gas holdup, a dimensionless key parameter for design purposes characterizing the transport phenomena of bubble column systems, defined as the volume fraction occupied by the gas phase. Many different methods are available for determining the gas holdup in a bubble column. The gas holdup parameter can be measured using techniques such as the increase in fluid height when gas is introduced (height difference method), dynamic gas disengagement technique, pressure drop measurements, electric resistivity probes, ultrasonics, computed tomographic scans, gamma-ray densitometry, and electrical resistance tomography. Some of these methods provide an overall gas holdup parameter, such as the height difference method, while others allow for examining the distribution of the gas holdup parameter at certain locations along the tower (axial and radial distributions), such as the pressure difference and electrical resistance tomography methods, in addition to providing an overall gas holdup measurement. These methods are advantageous because it allows one to determine the non-uniformities in reactor performance. In general, gas holdup is known to increase with increasing gas velocity, operating pressure, and with the addition of surface active agents (surfactant); whereas it decreases with increasing liquid viscosity and solids concentration. In smaller bubble columns of diameter greater than 10 - 15 cm and height above 1 - 3 m (or aspect ratio (height to diameter ratio) greater than 5), there is negligible effect on the gas holdup. At low gas superficial velocities, the gas holdup is dependent on the design of the gas distributor (number of holes, pitch, and diameter of the orifice holes) (Perry and Green, 1997; Kantarci *et al.*, 2005).

Gas-liquid mass transfer is an important process in many chemical engineering unit operations. Many models exist to describe and predict the mass transfer in two phase systems; however, mass transfer models in slurry bubble systems are limited due to their complicated reaction system between the gas, liquid, and solid phases.

The volumetric mass transfer coefficient, $k_L a$, is an important parameter that quantifies the reactor performance. Measurement methods include: dynamic pressure step method (DPM), steady-state physical method (SPM), classic sulfite method (CSM), steady-state sulfite method (SSM) (Havelka *et al.*, 2000), oxygen desorption method using a high pressure optical fibre oxygen probe (Lau *et al.*, 2004), dynamic absorption technique by gas chromatography (Jin *et al.*, 2004), and the dynamic oxygen absorption technique using an oxygen probe (Letzel *et al.*, 1999; Rewatkar and Bennington, 2004; Vandu *et al.*, 2004; Vandu and Krishna, 2004), to name a few.

In general, the volumetric mass transfer coefficient increases with gas velocity, gas density, pressure, and the presence of surfactants (due to the creation of smaller bubbles) but decreases with increasing solids concentration and liquid viscosity (Perry and Green, 1997; Kantarci *et al.*, 2005).

4.2 Experimental Apparatus

The experimental apparatus (Figure 4.1) was designed and built to simulate industrial pulp retention towers. It consists of a cylindrical tubular vessel which was built with a 1.27 cm (1/2-inch) thick Plexiglas wall and an inside diameter of 27.94 cm (11 inches). The total height of the cylindrical column is 3.26 metres and is made up of five individual sections, labelled, A1, A2, B, C1, C2, and D with 2.54 cm thick flanges at both ends of each section for fastening with screws and bolts. The top flange of each section contains a groove to fit a rubber o-ring for creating a good seal between each of the sections in order to prevent fluid leakage. The apparatus may be configured so that it may operate in batch or continuous mode (with the addition of a downcomer pipe returning to the tank). However, this study is limited to the batch configuration.

In the batch-operated case, a gas distributor plate is installed at the bottom of the column. The distributor plate consists of 400 - 1 mm diameter holes uniformly distributed in the 2.54 cm thick circular plate to ensure uniform distribution of the gas phase. For more information on the design of the gas distributor plate please refer to Pineault (1999). Air is introduced to the column via a compressed air line and passes through the distributor plate and into the column where it makes contact with the fluid inside the vessel.



Figure 4.1: Schematic of cylindrical experimental apparatus for batch operation

The tower is equipped with four pressure transducers, (Cole Parmer model number 07356-00, Vernon Hills, IL), with 1 to 5 volts DC output and a pressure range of 0 to 15 psig. Pressure transducers are used to monitor the pressure inside the vessel in order to estimate the overall gas holdup characteristic. The use of pressure transducers does not require a transparent column or vessel and they may be applied in industrial reactors without compromising or disrupting the flow of material inside the vessel (Camarasa *et al.*, 1999).

In this study, the transducers are located at 40.2 cm, 110.5 cm, 180.7 cm, and 251.0 cm above the gas distributor and all transducers are aligned flush along the inside wall of the tower to eliminate any disturbance of the flow of the contents inside the vessel. The pressure transducers are connected to a computer where the pressure is monitored using the LabView data acquisition software system (National Instruments Corporation, Austin, TX).

A commercial electrical resistance tomography system model P2000 (Industrial Tomography Systems, Ltd., Manchester, UK), is also used for data collection. The sensor array consists of 8 planes of 16 equally-spaced peripheral electrodes mounted flush around the inside wall of the tower in a non-intrusive manner with regards to the flow of fluid inside the vessel. Figure 4.2 shows a schematic of the configuration of the sensors, Figure 4.3 shows a picture of the actual sensor electrode, and Figure 4.4 is a picture of the 16 electrode arrangement mounted on the tower with a 2% softwood pulp suspension inside.



Figure 4.2: Configuration of 16-electrode sensor arrangement (ITS, 2003)



Figure 4.3: Picture of electrode sensor



Figure 4.4: Portion of cylindrical tower with 2 planes of 16-electrode sensors in $C_m = 2\%$ SW

Pairs of electrode planes (1-2, 3-4, 5-6, 7-8) separated by 7.5 cm are equally spaced (64.4 cm intervals) along the tower height. The location of the 8 planes on the cylindrical tower is as follows: Plane 1 at 17.7 cm, Plane 2 at 25.2 cm, Plane 3 at 89.5, Plane 4 at 97.0 cm, Plane 5 at 161.4 cm, Plane 6 at 168.9 cm, Plane 7 at 233.2 cm, and Plane 8 at 240.7 cm. The electrode-sensor assembly material is made of 316 stainless steel. The assembly consists of a circular stainless steel sheet with a 2.19 cm diameter and 0.0254 cm thickness designed as per manufacturer's (ITS, Ltd., Manchester, UK) specifications (which is a function of the vessel diameter, range of conductivity to be measured, velocity of material flowing, and the required imaging speed). The stainless steel rod is 8.5 cm in length and 0.159 cm (1/16 inches) in diameter. The electrodesensor assembly is soldered together to make contact between them. One end of the electrical cables/wires are then attached to the rod end of each of the electrode-sensor assemblies and then to an IEEE 488 connector pin which connects to the ITS P2000 instrument. Four grounding electrodes positioned 15 centimetres above the upper plane of the pair of planes 1-2, 3-4, 5-6, 7-8 and in electrical contact with the fluid inside the vessel but mounted in a non-invasive manner ensures all voltage measurements are fixed to a common ground source complete the system. The grounding electrodes are made of 0.635 cm (1/4 inch) diameter stainless steel rods 10.16 cm (4 inches) in length with one end making electrical contact with the internal fluid and the other connected to the IEEE 488 connector pin via electrical cable as with all other electrode-sensors. The P2000 data acquisition system unit (ITS, Ltd., Manchester, UK) is connected to a laptop by means of an RS232 serial cable. The voltage measurements made using the ERT system have an accuracy of \pm 0.5% and repeatability of > 99.5%. The ERT system has excellent temporal resolution; however, spatial resolution is only about 5-10% of the vessel diameter (ITS, 2003).

The experimental apparatus also consists of a mass flow meter, air and nitrogen lines, a holding tank, and a pump, as shown in Figure 4.1.

4.3 Experimental Methods and Techniques

4.3.1 Pulp Preparation

Identical pulp source was used in the cylindrical tower experiments as in the rectangular channel experiments. Please refer to Chapter 3 for pulp fibre properties and characteristics. To prepare a pulp suspension of desired mass concentration, paper sheets were torn by hand, weighed, placed in buckets, and allowed to soak in water overnight. The mass of paper sheets used was determined based on the desired consistency for 150 L of suspension. The following equation was used to determine the consistency with the density of fibre and water taken as 1520 kg/m³ and 1000 kg/m³, respectively.

$$C_m = \frac{m_f}{m_f + m_w} * 100\%$$
(4.1)

Where C_m represents the pulp mass concentration, m_f is the mass of fibres, and m_w is the mass of water.

The soaked paper sheets were then placed in a repulper where the mechanical action of the repulper blades separated the fibres and provided a homogeneous pulp suspension. Once all of the paper sheets were repulped, the entire suspension was placed in the holding tank and filled with the remaining water to make up to 150 L and mixed for 5 - 10 minutes to ensure a uniform homogeneous suspension. The pulp suspension was then pumped into the tower and ready for experimental trials.

4.3.2 Experimental Preparation

In the batch-operated case, the initial suspension height was recorded. The conductivity of the suspension was measured using a conductivity meter. The background pressure readings were recorded to establish a pressure reference for the particular suspension in the vessel in order to determine the gas holdup using the pressure difference method.

The P2000 ERT unit is connected to a laptop by means of an RS232 serial cable and the power adaptor is connected to the mains supply. Each plane of electrodes on the ERT sensors that will be used in the tests is connected to the instrument by means of the 36-way connections on the front of the unit. The laptop is turned on, the ITS software is initiated, and finally, the P2000 instrument is turned on. A new test run is open and a setup screen appears for user configuration. Figure 4.5 shows an example of the basic configuration window. For more information on the configuration settings, please refer to ITS Ltd. (2003).

lame	Favoured cl	Favoured client						
escriptic)	Plane 1 test	with sensor si	mulator					
DAS S	ettings		- Data Collection					
Number	of planes	1	Maximum number of frames	0				
Samples	per frame	8 🔻	Sampling time interval (ms)	55				
Frames (per download	1	Electrical Properties					
Injection	current	15	Media conductivity (mS/cm)	1				
			Object conductivity (mS/cm)	0				
Advanced Configuration		Iration	Conversion for Non-conductive phase					

Figure 4.5: ITS P2000 basic configuration window (ITS, 2003)

Prior to data collection with the ERT system, a reference measurement is taken as a basis for the subsequent measurements. Since the ERT is being used to determine gas holdup, the reference measurement is taken when the column is filled with fluid only (i.e., no gas bubbles present). This reference measurement is called the reference frame. Air was introduced into the tower at the desired air flow rate (0.00272 to 0.0272 m/s). Prior to starting the data collection process, sufficient time was provided for the air to mix with the suspension. This process approximately took 2 - 5 minutes. Upon sufficient established mixing time, the data collection process was begun by initiating the pressure measurement data collection process which was accomplished using the LabView data collection program, starting the electrical resistance tomography data collection process and also measuring the bubbling height of the suspension to determine the gas holdup by the height difference method.

4.3.3 Gas Holdup Experiment

Gas holdup is a dimensionless parameter describing the volume fraction that is occupied by the gas-phase in a two- or three-phase system. Gas holdup is an important parameter because it is used in the design and analysis of bubble column studies to examine and describe the reactor performance.

Gas holdup was determined in the cylindrical bubble column by applying three different measurement techniques: the height difference method, the pressure difference method, and the electrical resistance tomography method. Tests were carried out with water and fully-bleached kraft pulp suspensions (identical pulp source used in the 2-D studies) with mass concentrations studied between 0.5 and 9%. The height difference method required visual measurements and data was recorded by hand. Data was collected by a PC connected to the pressure transducers for the pressure difference method and a laptop connected to the electrical resistance tomography unit was used for the ERT method. The data was then analyzed to determine the gas holdup for each respective method as discussed in the *Data Analysis* section.

4.3.4 Gas-Liquid Mass Transfer Experiment

Mass transfer experiments were conducted in the cylindrical column under batch operating conditions. Tests were carried out with water and fully-bleached kraft pulp suspensions having mass concentrations between 0.5 and 4%. The volumetric mass

transfer coefficient, $k_L a$, was determined using the dynamic oxygen absorption technique, a technique commonly used in literature (Letzel et al., 1999; Rewatkar and Bennington, 2004; Vandu et al., 2004; Vandu and Krishna, 2004).

The column, filled with the liquid phase, was first sparged with nitrogen gas in order to strip the dissolved oxygen present in the column to a negligible concentration near zero. After a negligible dissolved oxygen concentration was reached, air was introduced through the distributor and into the column at a specified flow rate (ranging from 0.00272 to 0.0272 m/s). The dissolved oxygen concentration was monitored as a function of time by a dissolved oxygen probe and meter model 53101-24 manufactured by Thermo Scientific Orion (Beverly, MA), placed at a height of 110.5 cm above the gas distributor (approximately at the midpoint of the gas-free suspension height) and inset 1 cm. This sampling method was applied to water and low consistency pulp suspensions $(C_m \le 2\% \text{ SW and } C_m \le 3\% \text{ HW})$. For pulp suspensions with higher mass concentrations, the sampling method was modified. Samples were withdrawn from a sampling port using a syringe, dispensed in a 25 mL Erlenmeyer flask, and then the dissolved oxygen concentration was measured. Every effort was made to minimize exposure of the contents of the flasks to the surrounding air by placing stoppers on the flasks as well as ensuring that there was no head-space present in the flasks between sampling and measurement steps.

4.4 Data Analysis

4.4.1 Gas Holdup

Gas holdup (ε_g) was determined by applying three different measurement techniques: height difference, pressure difference, and electrical resistance tomography.

4.4.1.1 Height Difference Method

The height difference method is a visual technique in which the height of the suspension under steady-state gas-dispersed conditions, H_b , is measured and compared to the theoretical gas-free pulp height, H_s . The presence of small bubbles may be trapped in the fibre networks; however, it has been found not to be more than a few percent even

under batch operating conditions and their effect on the overall gas holdup is negligible (Pelton and Piette, 1992). The theoretical gas-free pulp height, H_s , was used regardless in the calculation; however, it was found that indeed the theoretical gas-free pulp height and the measured gas-free pulp height were identical for all mass concentrations. The following equation is used to determine the gas holdup using the height difference method:

$$\varepsilon_g = \frac{H_b - H_s}{H_b} \tag{4.2}$$

The theoretical gas-free pulp height, H_s , is determined by knowing the mass of pulp in the tower as well as knowing the tower dimensions:

$$H_s = \frac{4m_p}{\rho_p \pi D_c^2} \tag{4.3}$$

where m_p is the mass of the pulp suspension, ρ_p is the density of the pulp suspension, and D_c is the internal column diameter. The density of the pulp suspension is calculated knowing the density of fibre, water, and consistency, using:

$$\rho_p = \frac{1}{\left(\frac{C_m}{\rho_f} + \frac{(1 - C_m)}{\rho_w}\right)} \tag{4.4}$$

where C_m is the mass fraction of fibre in suspension and defined as:

$$C_m = \frac{m_f}{m_f + m_w} \tag{4.5}$$

where m_f is the mass of fibres and m_w is the mass of water.

4.4.1.2 Pressure Difference Method

Gas holdup determination using pressure transducers is a widely used technique to determine gas holdup (Shah *et al.*, 1982; Vandu and Krishna, 2003; Hol and Heindel, 2005; Kantarci *et al.*, 2005). The technique is based on measuring the pressure at points along a tower and obtaining the pressure drop between successive transducers. The following equation is used to determine the gas holdup in the tower:

$$\varepsilon_g = 1 - \left(\frac{\Delta P}{\Delta P_o}\right) \tag{4.6}$$

Where ΔP is the pressure drop between two successive pressure transducers when the tower is gassed and ΔP_o is the corresponding pressure drop when the tower is un-gassed and is determined from the suspension hydrostatic head. This method can be applied to determine the gas holdup at points between two transducers or an average may be taken to determine the overall gas holdup of the entire tower.

4.4.1.3 Electrical Resistance Tomography Method

The third and final measurement technique to determine the gas holdup is the electrical resistance tomography method. Using the conductivity data obtained from the ERT system, the gas holdup is determined by applying the Maxwell equation which is suitable for systems with spheres or spherical-like particles distributed in a continuous phase:

$$\varepsilon_{g} = \frac{2\sigma_{1} + \sigma_{2} - 2\sigma_{mc} - \frac{\sigma_{mc}\sigma_{2}}{\sigma_{1}}}{\sigma_{mc} - \frac{\sigma_{2}}{\sigma_{1}}\sigma_{mc} + 2(\sigma_{1} - \sigma_{2})}$$
(4.7)

where σ_1 and σ_2 are the conductivities of the continuous and dispersed phases, respectively in mS/cm, and σ_{mc} is the conductivity of the reconstructed measured conductivity by the ERT system. When a non-conductive phase is present, such as gas bubbles in liquid, the Maxwell equation is further simplified to:

$$\varepsilon_g = \frac{2\sigma_1 - 2\sigma_{mc}}{\sigma_{mc} + 2\sigma_1} \tag{4.8}$$

The conductivity of the continuous phase, σ_l , which is the conductivity of the pulp suspension or water, can be measured using a conductivity meter and the conductivity of the dispersed phase, σ_2 , is zero because it represents the non-conductive gas phase.

4.4.2 Mass Transfer

The rate at which mass transfer occurs depends mostly on factors that affect the interfacial area, a. In gas-liquid reactors, as in the oxygen delignification reactor for example, the mass transfer from the gas phase to the liquid phase is the most important goal of the process. Prediction of $k_L a$ is an important part of gas-liquid reactor design since it is an indication of overall reactor performance. Factors that may influence the gas-liquid mass transfer include temperature, pressure, viscosity, density, surface tension, surfactants, solids concentration, aeration velocity, and pH, to name a few.

Gas-liquid mass transfer between oxygen and the water phase of the pulp fibre suspension is often characterized using the liquid-side mass transfer coefficient, k_L (m/s). However, due to the difficulty in measuring this parameter directly, the characterization is further defined by the "lumped" term known as the volumetric mass transfer coefficient, $k_L a$, and *a* is the gas-liquid interfacial area per unit dispersion volume (m²/m³) which can be measured more easily. $k_L a$ was determined using:

$$\frac{dC}{dt} = k_L a(C^* - C) \tag{4.9}$$

where C is the concentration of dissolved oxygen and C^* is the saturated oxygen concentration. Since the dissolved oxygen probe has a finite response time, a sensor

constant that quantifies the response time of the probe was determined. Two separate beakers containing the liquid, one saturated with nitrogen and one saturated with air were used. The probe was placed in the nitrogen-saturated beaker until a negligible dissolved oxygen concentration was reached and then placed immediately in the air-saturated beaker. The dissolved oxygen concentration was measured as a function of time and k_s (the sensor time constant) calculated using equation 4.10. Using this method, it was assumed that the liquid was perfectly mixed and the depletion of the oxygen from the gas bubble was negligible.

$$\frac{dC_s}{dt} = k_s (C^* - C_s) \tag{4.10}$$

Integrating equation 4.10 gives:

$$\frac{C_s}{C^*} = 1 - e^{-k_s t} \tag{4.11}$$

The sensor equation resulting from the finite response time of the dissolved oxygen probe is given by:

$$\frac{dC_s}{dt} = k_s(C - C_s) \tag{4.12}$$

Equations 4.9 and 4.12 are solved simultaneously to yield:

$$\frac{C_s}{C^*} = 1 - \frac{1}{k_s - k_L a} \left[k_s e^{-k_L a t} - k_L a e^{-k_s t} \right]$$
(4.13)

Finally, $k_L a$ was determined from time resolved measurements of dissolved oxygen concentration and using the solver function in Excel. k_s was determined in separate tests to be 0.069 ± 0.015 s⁻¹ in water and 0.031 ± 0.0083 s⁻¹ in low-consistency ($C_m = 0.5$ to 4%) pulp suspensions. Probe response time was rapid compared with the gas-liquid mass

transfer rates measured and did not affect the $k_L a$ values computed. This technique has been well established (Letzel *et al.*, 1999; Vandu *et al.*, 2004(a); Han and Al-Dahhan, 2007).

4.5 Results and Discussion

4.5.1 Gas Holdup

Two of the methods applied to determine the gas holdup, height difference and pressure difference methods were used to compare and establish the validity of the third method's (ERT) ability to accurately determine the gas holdup. Previous studies (Jin, Wang, and Williams, 2007; Vijayan, Schlaberg, and Wang, 2007; Dong *et al.*, 2005; Fransolet, *et al.*, 2005; Fransolet *et al.*, 2001) have applied the electrical resistance tomography method to determine gas holdup and bubble characteristics in two- or three-phase media; however, no literature has been found to-date regarding pulp suspensions and applying the electrical resistance tomography method in determining gas holdup. The advantage to the electrical resistance tomography method is that in addition to estimating the overall gas holdup in a vessel, it also provides insight into the radial distribution, a parameter that the height difference and pressure difference methods cannot.

In this section, a discussion of the results obtained for the gas holdup from all three measurement techniques is presented. The results are presented for water, saltwater, and hardwood and softwood pulp suspensions of mass concentrations between 0.5% and 9%. The gas holdup parameter was determined typically to within 2% for all three measurement techniques and a repeatability of $\pm 0.1\%$ for the height difference method, $\pm 0.5\%$ for the pressure difference method, and $\pm 0.2\%$ for the ERT method in a single test for $C_m = 4\%$ hardwood pulp as an example.

4.5.1.1 Water and Saltwater Results

Experiments were conducted in water and saltwater solutions of concentrations 0.1 g/L, 0.5 g/L, and 1 g/L, using the batch-operated bubble column configuration. The saltwater solution trials were conducted in an effort to obtain greater signal-to-noise ratio (SNR) for the ERT method. For example, the conductivity of tap water was determined to be between 0.017 and 0.018 mS/cm, but with the addition of salt, a salt solution of 0.1 g/L produces a conductivity of 0.2 mS/cm, which greatly improves the SNR of the ERT system from 35 dB to 62 dB (Stephenson, York, and Mann, 2007). Figures 4.6 (a) and 4.6 (b) show the results of the gas holdup for water and a 0.1 g/L saltwater solution, respectively, as a function of superficial air velocity with all three measurement techniques. In all cases, the gas holdup increases with superficial gas velocity. For superficial gas velocities between 0.00272 m/s to 0.0272 m/s the gas holdup ranges from 0.6% to 17%. The three methods are almost identical with the slight addition of salt compared to the water case alone. The addition of salt aids in the reconstruction image from the ERT measurements since the measurements are based on the conductivity of the fluid compared to the non-conductive phase. Figures B1.2, B1.3, and B1.4 in Appendix B1 presents additional experiments with different concentrations of saltwater solutions.



(b)

Figure 4.6: Gas holdup versus superficial gas velocity in (a) water, and (b) 0.1 g/L saltwater solution with three different measurement techniques

Figure 4.7 shows a comparison between the experimental height method of determining gas holdup along with literature data. Data from Rewatkar and Bennington (2004) had been taken using a geometrically identical apparatus (inside diameter of 27.94 cm) and gas distributor (distributor plate containing 400 1 – mm diameter holes), as used in this study, along with the height method for determining the gas holdup. Hol and Heindel (2005) measured the gas holdup in a 32.1 cm inside diameter vessel with 486 1 – mm diameter holes in the distributor using the pressure difference method. Reese *et al.* (1996) applied the height method for gas holdup determination in a 10.2 cm internal diameter and 2.2 m high bubble column with uniform gas distribution. The relation between the experimental results of this study and literature shows the same trend; the gas holdup increases with superficial gas velocity, but differences in the distributor design and vessel dimensions have an effect on the magnitude of the gas holdup result.



Figure 4.7: Overall gas holdup versus superficial gas velocity for water

Many correlations exist in the literature to predict the gas holdup in bubble columns and slurry bubble columns. It is reported that the main factors affecting the gas holdup include: superficial gas velocity, liquid and solid phase properties, column dimensions, operating conditions (temperature and pressure), and gas distributor design. However, few correlations exist for systems involving pulp suspensions.

4.5.1.2 Pulp Suspension Results

Figures B1.5 through B1.11 in Appendix B1 presents the gas holdup as a function of superficial gas velocity for hardwood pulp suspensions of mass concentrations ranging from 0.5% to 9% measured using all three measuring techniques. Similarly, Figures B1.12 through B1.16 in Appendix B1 is for softwood pulp suspensions of mass concentrations ranging from 0.8% to 4%. Figure 4.8 presents the results of the gas holdup for $C_m = 3\%$ hardwood pulp suspension as a function of the superficial gas velocity with all three measurement techniques (the height method, the pressure difference method, and the ERT method). The three methods agree well with each other with a maximum standard deviation of 0.2%. The gas holdup increases with the superficial gas velocity but the magnitude of the gas holdup is much lower compared to the water and saltwater cases (Figure 4.6) at the same superficial gas velocity.



Figure 4.8: Gas holdup versus superficial gas velocity in $C_m = 3\%$ HW with three different measurement techniques

Figures 4.9 (a) and (b) presents the gas holdup as a function of pulp mass concentration for hardwood and softwood pulps, respectively, using the height difference method. As the concentration of fibres increases in the pulp suspension, the gas holdup decreases and the results are comparable to literature results for pulp suspension of < 2% mass concentration (Walmsley, 1992; Reese *et al.*, 1996; Su and Heindel, 2003; Xie *et al.*, 2003; Hol and Heindel, 2005). The gas holdup decreased as the fibre mass concentration increased because of the increase in resistance to gas flow due to the presence of fibres. The increase in fibre presence causes the bubbles to coalesce, forming larger-sized bubbles which rise faster up through the stationary fluid. As the bubble rise velocity increases, the bubble retention time decreases, resulting in a decrease in gas holdup. This was validated by the visual observations made in the cylindrical column but also in the rectangular channel where individual bubbles in suspension were analyzed for bubble shape, size, and rise velocity, as discussed in Chapter 3.



(b)

Figure 4.9: Overall gas holdup versus kraft pulp mass concentration (a) hardwood (b) softwood

The presence of gas channelling was observed near the bottom of the tower near the gas distributor at low superficial gas velocities and for pulp mass concentrations as low as 2% in softwood pulp. Hol and Heindel (2005) also observed channelling for pulp suspensions of mass concentrations as low as 0.8% and for low superficial gas velocities. However, the channelling effect decreased or was non-existent as the superficial gas velocity increased or the gas velocity was sufficiently high enough such that it was no longer present.

4.5.1.3 Axial Distribution Results

The gas holdup axial distribution was determined using the gas holdup results from the pressure difference and ERT methods. The pressure difference method results in local gas holdup locations along the height of the tower, and the ERT method allows for gas holdup results at 6 locations along the height of the tower corresponding to the locations of the 6 ERT planes.

Figures B2.1 through B2.16 in Appendix B2 shows the relationship between the axial height and the gas holdup results with superficial gas velocities ranging 0.00272 m/s to 0.0272 m/s for water and varying pulp mass concentrations.

Figures 4.10 and 4.11 presents the gas holdup axial distribution for water and 0.1 g/L saltwater solution, respectively, with varying superficial gas velocities. The pressure difference method and ERT methods agree at low superficial gas velocities (i.e. $U_g \leq 0.0068 \text{ m/s}$), but the gas holdup axial distribution for the two methods deviate at higher superficial gas velocities (i.e. $U_g > 0.0068 \text{ m/s}$) and the deviation is greater in the water case. This was also observed and described earlier for the overall gas holdup results determined from all three measurement techniques. The gas holdup values are consistent with the axial height at low superficial gas velocities ($U_g \leq 0.0068 \text{ m/s}$), but as the superficial gas velocity increases, variation of the gas holdup along the tower height is observed, mostly due to recirculation of the fluid causing the bubbles to coalesce and rise faster up the tower, decreasing the gas holdup results in some cases, or forming channels for the bubbles to pass resulting in higher gas holdup values at certain locations. This

phenomenon occurred mostly in cases with high pulp mass concentrations, example, for 4% softwood and 9% hardwood.



Figure 4.10: Gas holdup axial distribution for water with varying superficial gas velocity measured using the pressure difference method (solid symbols) and the ERT method (open symbols)



Figure 4.11: Gas holdup axial distribution for 0.1 g/L saltwater with varying superficial gas velocity measured using the pressure difference method (solid symbols) and the ERT method (open symbols)

Figures 4.12 and 4.13 shows the relationship between the axial height and the gas holdup results with varying pulp mass concentrations at a fixed superficial gas velocity.

Hol and Heindel (2005) examined the axial distribution of the gas holdup at a fixed gas superficial velocity ($U_g = 10 \text{ cm/s}$) and found that for pulp mass concentrations of softwood fibres ranging 0.05 to 1.8% there was no significant effect on the axial variation of local gas holdup. A local gas holdup maximum was observed for all trials and was consistently at or near the same axial position. For a fixed axial location, the local and overall gas holdup decreased as the pulp mass concentration increased due to increased bubble coalescence.



Figure 4.12: Gas holdup axial distribution for varying mass concentrations of hardwood pulp and a fixed superficial gas velocity ($U_g = 0.014 \text{ m/s}$) measured using the pressure difference method (solid symbols) and the ERT method (open symbols)



Figure 4.13: Gas holdup axial distribution for varying mass concentrations of softwood pulp and a fixed superficial gas velocity ($U_g = 0.014$ m/s) measured using the pressure difference method (solid symbols) and the ERT method (open symbols)

4.5.1.4 Radial Distribution Results

The radial distribution of the gas holdup was determined using the data obtained from the ERT unit. The conductivity data was averaged over 5 sections of the crosssection of the vessel and then converted into gas holdup data as described in the *Data Analysis* section. Figure 4.14 shows the zones that were used to isolate conductivity data for further analysis. Average conductivity data of zones 2, 6, 11, 16, and 20 were used to report the average radial gas holdup.



Figure 4.14: ERT regionization zones for zone average data

In water and at low superficial gas velocity, the gas holdup is higher on one side of the tower, but as the bubbles rose higher up the tower, the bubbles were more evenly distributed, resulting in a more uniform radial distribution (see Figure 4.15). Figure 4.16 shows the gas holdup radial distribution by planes for water with varying superficial gas velocities.



Figure 4.15: Gas holdup radial distribution by planes measured using ERT for water at a fixed superficial gas velocity (0.0027 m/s)



Figure 4.16: Gas holdup radial distribution by planes measured using ERT for water at various superficial gas velocities


Figure 4.17: Gas holdup radial distribution by planes measured using ERT for 1% hardwood pulp at various superficial gas velocities

Lindsay *et al.* (1995) measured the radial gas holdup in water, 1% and 2% southern softwood pulps using γ -densitometry. Results in water showed that for gas superficial velocities less than 0.0263 m/s, corresponding to the bubbly flow regime, the gas holdup was a linear function of the superficial gas velocity and the distribution was relatively uniform and constant along the height of the tower. In this study, visual observations confirmed that as soon as the gas bubbles disengaged from the distributor, the bubbles tend to flow towards one side of the tower, but then distributed more evenly as the bubbles rose higher up the tower. This is due to the fact that as the gas is first introduced at low superficial velocity to the column filled with liquid, the internal recirculation of the liquid forces the bubbles to flow to one side, but at higher superficial velocities, the gas bubbles have enough momentum to flow uniformly up the tower without being forced to one location over another.

In most cases, the gas holdup is greater near the centre of the tower and lower near the tower walls. This is comparable to literature investigations with water and low consistency pulp suspensions (Lindsay *et al.*, 1995; Xie *et al.*, 2003; Camarasa *et al.*, 1999; Schulz and Heindel, 2000; Vijayan *et al.*, 2006). However, in this study, when the fibre mass concentration was increased, for example in the case of 6% hardwood and 4% softwood (see Figures 4.18 and 4.19), the distribution deviated from uniformity with higher gas holdup occurring near one side of the tower walls in some cases due to the increase in fibre presence restricting the flow of the gas phase. Literature results were not found for higher consistency pulp suspensions (greater than 2%).



Figure 4.18: Gas holdup radial distribution measured using ERT for plane 3 and $U_g = 0.020$ m/s for various hardwood mass concentrations



Figure 4.19: Gas holdup radial distribution measured using ERT for plane 3 and $U_g = 0.020$ m/s for various softwood mass concentrations

A complete set of graphs pertaining to the radial distribution can be found in Appendix B3.1 through B3.70 in Appendix B3 showing the radial distribution for water, saltwater, hardwood and softwood pulp suspensions at various mass concentrations and superficial gas velocities for planes 1 through 6.

4.5.2 Mass Transfer

In this section, a discussion of the results obtained for the mass transfer in the batch-operated bubble column is presented. The results are presented for water and hardwood and softwood pulp suspensions of mass concentrations between 0.5% and 4%.

The volumetric gas-liquid mass transfer coefficient was determined in water and pulp suspensions of varying mass concentrations ($C_m = 0.5 - 4\%$). $k_L a$ was determined typically to within 9% in a single test with a repeatability of ±12% for multiple determinations. The experimental data for the volumetric mass transfer coefficient obtained for water was compared to the data found in literature and is presented in Figure

4.20. As the superficial gas velocity was increased, the volumetric mass transfer coefficient increased as well.



Figure 4.20: Volumetric mass transfer coefficient versus superficial gas velocity in water with literature correlations

The experimental results obtained in this study are similar to those obtained by Rewatkar and Bennington (2004) but are higher than other published results. This difference could be due to the differences in vessel dimensions and method of air introduction since it is known that the volumetric mass transfer coefficients depend on the gas velocity, reactor geometry, sparger design, and presence of solid particles, especially those that promote or prevent coalescence (Shah *et al.*, 1982; Kantarci *et al.*, 2005). The volumetric mass transfer values obtained in this study matched more closely to Rewatkar's data since the vessel dimensions and method of air introduction were identical. In the study of Shimizu *et al.* (2000) the vessel dimensions were 0.20 m in diameter with a ring sparger containing 12 holes of 1.0-mm diameter. Shah *et al.* (1982) reported the correlation for a bubble column of 0.20 m in diameter with a cross of nozzles 1.0 mm in diameter.

In this study, the internal column diameter is 27.94 cm with 400 - 1 mm diameter holes uniformly distributed. Figures 4.21 and 4.22 shows the experimental k_La results plotted versus consistency, with superficial gas velocity as a parameter for softwood and hardwood pulps, respectively, along with literature results for the softwood pulp case.



Figure 4.21: $k_L a$ versus percent mass concentration of bleached softwood kraft pulp as a function of superficial gas velocity



Figure 4.22: $k_L a$ versus percent mass concentration of bleached hardwood kraft pulp as a function of superficial gas velocity

The trends for both pulps are similar. The volumetric mass transfer coefficient of hardwood pulps at equivalent consistencies are on average slightly greater than that of softwood pulps. Both pulps show a decrease in $k_L a$ as the consistency of the pulp suspension increases. The effect of the superficial gas velocity is depicted well in the hardwood case. As the superficial gas velocity increases, so too does the mass transfer coefficient, with higher mass transfer rates exhibited at higher superficial gas velocities as one would expect. At pulp mass concentrations greater than 2% (SW) and 3% (HW), the volumetric mass transfer coefficient collapses to $k_L a$ between 5*E*-4 and 1*E*-3 s⁻¹ for all superficial gas velocities. This can be attributed to the formation of larger gas bubbles rising up faster in the suspension or due the formation of gas channels, decreasing the interfacial area between the bubble and the fibre suspension and thereby decreasing the volumetric mass transfer rates.

The volumetric mass transfer coefficient is affected by both the superficial gas velocity and the mass concentration of the pulp suspension. At low superficial gas velocities and low mass concentrations of pulp suspension, less gas is present with bubbles rising slowly up through the suspension resulting in low $k_L a$ values. At higher superficial gas velocity with low mass concentration of pulp suspension, more gas is present in the column for better mass transfer between the pulp fibres and the gas phase, resulting in higher $k_L a$ values at low consistency. However with a higher mass concentration of pulp suspension, even at high superficial gas velocities, the mass transfer ability is diminished. The network strength of the suspension is so great that it causes the bubbles to either coalesce into large slugs of bubbles (mostly observed in the hardwood pulp case) or causes the gas phase to channel along the sides of the walls of the tower (mostly observed in the softwood pulp case), resulting in a non-uniform distribution of the phases, lowering the gas-liquid interfacial area and thereby decreasing the $k_L a$ values. Also, the fact that the dissolved oxygen probe was positioned at one specific location (with samples being taken at the same location in cases where the dynamic method was not possible) may affect the mass transfer characteristic as well. If the channelling were to occur near the dissolved oxygen probe, then an increase in the $k_L a$ values would have been observed due to the localized presence of the gas phase in the suspension. The bubble shape, size, and rise velocity data obtained with the rectangular vessel along with Figures 4.8(a) and 4.8(b), showing the gas holdup results versus mass concentration for hardwood and softwood pulps, validates the mass transfer results obtained. The gas holdup in the tower is high at higher aeration rates but decreases as the mass concentration increases, no matter what aeration rate, reducing to 3.5% gas holdup in hardwood and 3% gas holdup in softwood at 4% mass concentration in both cases from a high of 13% holdup in water.

CONCLUSIONS AND RECOMMENDATIONS

The objectives of this thesis were to observe and determine the characteristics of air bubbles in water and kraft pulp suspensions having mass concentrations between 1 and 7% in a rectangular vessel, to describe the hydrodynamic behaviour, in terms of the gas holdup, of the gas phase in water and kraft pulp suspensions having mass concentrations between 0.5 and 9%, and to determine the volumetric gas-liquid mass transfer characteristics using air with water and kraft pulp suspensions having mass concentrations between 0.5 and 4% in a batch-operated bubble column. All of the objectives of this thesis have been achieved and the results can be summarized with the following conclusions:

- 1. The shape and size of bubbles in a rectangular channel varied with air flow rate. In general, bubble size increased as the air flow rate increased in water and for most pulp mass concentrations. Bubble shape and size were significantly affected by the mass concentration of pulp fibres and the pulp type (hardwood or softwood at the same mass concentrations). Channelling of the gas phase was observed for pulp mass concentrations as low as $C_m = 2\%$ in softwood pulp and $C_m = 7\%$ in hardwood pulp. It was concluded that the inherent fibre characteristics attributed to this observation.
- 2. Gas holdup increased as the superficial gas velocity increased and the results were comparable to literature studies in water and low consistency softwood pulp suspensions. Gas holdup was significantly affected by the presence of pulp fibres with a dramatic decrease and subsequent levelling-off at pulp mass concentrations of $C_m = 1\%$ for softwood pulp and $C_m = 2\%$ for hardwood pulp at a fixed superficial gas velocity. The increase in fibre presence caused the bubbles to coalesce forming larger-sized bubbles which rose faster up through the fluid in the tower. The axial gas holdup distribution was consistent for all superficial gas velocities with results varying at most by about 1% for water, softwood and hardwood pulp suspensions. The radial gas

holdup distribution results showed that the gas holdup was predominatly greater near the centre of the tower and lower near the walls of the tower for all cases and this phenomenon was more pronounced as the superficial gas velocity increased. Results were comparable to literature studies with water and low consistency pulp suspensions; however, literature results were not found for cases with pulp suspensions of mass concentration greater than $C_m = 2\%$.

3. Volumetric mass transfer coefficient $(k_L a)$ in water increased as the superficial gas velocity increased and was comparable to literature results. $k_L a$ decreased with increasing pulp fibre presence which could be attributed to earlier observations that resulted in bubble coalescence and an increase in bubble size thereby reducing the interfacial area, *a*, and ultimately $k_L a$.

More research is required in order to understand the hydrodynamic and mass transfer characteristics of oxygen in pulp as in commercial pulp retention towers. Most of the research efforts have been focussed on batch situations with multiphase fluids other than pulp suspensions; and even fewer studies were found with co-current flow situations with multiphase fluids. However, pulp suspensions have a very unique flow characteristics that cannot be correlated to most multiphase systems; therefore, a unique study tailoring to the specific conditions found in industrial oxygen delignification systems is required.

FUTURE WORK

Recommendations for future work in the investigation of the gas hydrodynamics and mass transfer in pulp retention towers are:

- 1. To investigate the gas hydrodynamics in medium consistency pulp suspensions greater than those used in this study (softwood $C_m > 4\%$ and hardwood $C_m > 9\%$).
- 2. To investigate and apply an alternate method to accurately measure the mass transfer in pulp suspensions of mass concentrations greater than $C_m = 4\%$. In this study it was difficult to obtain these results due to phase segregation at higher mass concentrations near the oxygen probe.
- 3. To study the gas hydrodynamics and mass transfer in co-current flow operation with side-entering flow and with bottom-entering flow (by replacing the gas distributor with a solid Plexiglas plate having a 2-inch diameter hole in the centre). These results will provide a better understanding of the performance of industrial pulp retention towers which operate in co-current flow and do not benefit from the excellent gas distribution at the base of the tower.
- 4. To study the gas hydrodynamics and mass transfer in co-current flow operation with a conical-shaped entrance region with appropriate modifications to the ERT array of planes 1 and 2. This can be accomplished by replacing section D (in Figure 4.1) with a newly designed conical-shaped section. This study will provide a greater understanding into the performance in industrial systems since the industrial towers have conical-shaped entrance regions and can be compared to the results with the flat plate bottom (as described in "1").
- 5. To study the effect of gas dispersion (using a gas mixer prior to entering the tower) on the gas holdup and mass transfer characteristic.

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Appendix A1

2D Pictures of Bubbles Rising in Water and Pulp Suspensions



(a) (b) Figure A1.1: Bubbles in water (a) air flow rate = 1.6 L/min, (b) air flow rate = 3.2 L/min



(a) (b) Figure A1.2: Bubbles in water (a) air flow rate = 4.8 L/min, (b) air flow rate = 6.3 L/min





(a) (b) Figure A1.4: Bubbles in 1% softwood kraft pulp (a) air flow rate = 1.5 L/min, (b) air flow rate = 3.0 L/min



(a) (b) Figure A1.5: Bubbles in 1% softwood kraft pulp (a) air flow rate = 4.5 L/min, (b) air flow rate = 6.0 L/min



(a) (b) Figure A1.6: Bubbles in 1% softwood kraft pulp (a) air flow rate = 8.0 L/min, (b) air flow rate = 10.0 L/min



(a) (b) Figure A1.7: Bubbles in 2% softwood kraft pulp (a) air flow rate = 1.4 L/min, (b) air flow rate = 2.9 L/min



(a) (b) Figure A1.8: Bubbles in 2% softwood kraft pulp (a) air flow rate = 4.7 L/min, (b) air flow rate = 6.6 L/min



(a) (b) Figure A1.9: Bubbles in 2% softwood kraft pulp (a) air flow rate = 8.3 L/min, (b) air flow rate = 9.9 L/min



(a) (b) Figure A1.10: Bubbles in 4% softwood kraft pulp (a) air flow rate = 1.5 L/min, (b) air flow rate = 2.9 L/min



(a) (b) Figure A1.11: Bubbles in 4% softwood kraft pulp (a) air flow rate = 4.5 L/min, (b) air flow rate = 6.1 L/min



(a) (b) Figure A1.12: Bubbles in 4% softwood kraft pulp (a) air flow rate = 8.2 L/min, (b) air flow rate = 10.0 L/min



(a) (b) Figure A1.13: Bubbles in 1% hardwood kraft pulp (a) air flow rate = 1.6 L/min, (b) air flow rate = 3.6 L/min



(a) (b) Figure A1.14: Bubbles in 1% hardwood kraft pulp (a) air flow rate = 5.5 L/min, (b) air flow rate = 7.2 L/min



(a) (b) Figure A1.15: Bubbles in 1% hardwood kraft pulp (a) air flow rate = 8.4 L/min, (b) air flow rate = 10.1 L/min



(a) (b) Figure A1.16: Bubbles in 1% hardwood kraft pulp (a) air flow rate = 12.5 L/min, (b) air flow rate = 15.9 L/min



Figure A1.17: Bubbles in 1% hardwood kraft pulp with air flow rate = 19.4 L/min



(a) (b) Figure A1.18: Bubbles in 2% hardwood kraft pulp (a) air flow rate = 1.3 L/min, (b) air flow rate = 3.6 L/min



(a) (b) Figure A1.19: Bubbles in 2% hardwood kraft pulp (a) air flow rate = 5.7 L/min, (b) air flow rate = 7.7 L/min



(a) (b) Figure A1.20: Bubbles in 2% hardwood kraft pulp (a) air flow rate = 9.3 L/min, (b) air flow rate = 10.5 L/min



(a) (b) Figure A1.21: Bubbles in 2% hardwood kraft pulp (a) air flow rate = 12.6 L/min, (b) air flow rate = 15.2 L/min



Figure A1.22: Bubbles in 2% hardwood kraft pulp with air flow rate = 20.0 L/min



(a) (b) Figure A1.23: Bubbles in 4% hardwood kraft pulp (a) air flow rate = 1.2 L/min, (b) air flow rate = 4.0 L/min



(a) (b) Figure A1.24: Bubbles in 4% hardwood kraft pulp (a) air flow rate = 5.9 L/min, (b) air flow rate = 7.4 L/min



(a) (b) Figure A1.25: Bubbles in 4% hardwood kraft pulp (a) air flow rate = 9.1 L/min, (b) air flow rate = 11.8 L/min



(a) (b) Figure A1.26: Bubbles in 4% hardwood kraft pulp (a) air flow rate = 13 L/min, (b) air flow rate = 16 L/min



Figure A1.27: Bubbles in 4% hardwood kraft pulp with air flow rate = 19.1 L/min

Appendix A2

Bubble Rise Velocity and Shape Data

Media	Air Flowrate	Mean Bubble Rise Velocity		
	L/min	m/s		
	1.50	0.583		
Water	3.00	0.758		
	4.50	0.790		
	6.00	0.769		
	8.00	0.822		
	10.0	0 990		
	1.50	0.242		
	3.00	0.353		
000 0 -40/	4.50	0.420		
SW $C_m = 1\%$	6.00	0.543		
	8.00	0.630		
	10.0	0.658		
	1.40	0.444		
	4.50	0.500		
	6.00	0.543		
SW C=2%	6.60	0.500		
0 0 m -270	8.00	0.550		
	8.30	0.608		
	9.90	0.664		
	10.0	0.514		
	1.50	No bubbles		
	2.90	No bubbles		
SW C _m =4%	4.50	No bubbles		
	6.10	No bubbles		
	8.20	No bubbles		
	10.0	No bubbles		
	1.60	0.500		
	3.60	0.007		
HW C _m =1%	5.50	0.769		
	7.20	0.038		
	10.40	1 000		
	12.5	1 154		
	15.9	1.429		
	19.4	1 500		

Table A2.1: Bubble rise velocity data

Media	dia Air Flowrate Mean Bubble Rise Velocity		
	L/min	m/s	
	1.30	0.319	
	3.60	0.395	
	5.70	0.556	
	7.70	0.600	
HW <i>C_m</i> =2%	9.30	0.682	
	10.5	0.789	
	12.6	0.789	
	15.2	1.071	
	20.0	1.000	
	1.20	0.417	
	4.00	0.536	
	5.90	0.577	
	7.40	0.600	
HW C _m =4%	9.10	0.682	
	11.8	0.750	
	13.0	0.714	
	16.0	0.714	
	19.1	0.882	
	1.50	No bubbles	
	3.50	No bubbles	
HW C _m =7%	6.80	No bubbles	
	10.6	No bubbles	
	14.9	No bubbles	

Table A2.2: Bubble rise velocity data, equivalent diameter and dimensionless numbers

Media	Air Flowrate	Bubble Rise Velocity	D _e	Eo	М	Log(M)	Re
	L/min	m/s	m				
Water	1.50	0.583	0.0194	5.02E+01	2.5356E-11	-10.60	1.13E+04
	3.00	0.758	0.0308	1.27E+02	2.5356E-11	-10.60	2.33E+04
	4.50	0.790	0.0308	1.27E+02	2.5356E-11	-10.60	2.42E+04
	6.00	0.769	0.0333	1.49E+02	2.5356E-11	-10.60	2.56E+04
	8.00	0.822	0.0398	2.12E+02	2.5356E-11	-10.60	3.26E+04
	10.0	0.990	0.0424	2.41E+02	2.5356E-11	-10.60	4.19E+04

		Measured	Predicted	
Media	Air Flowrate	Bubble Rise Velocity	Bubble Rise Velocity	Percent Error
	L/min	m/s	m/s	%
Water	1.50	0.583	0.362	37.8
	3.00	0.758	0.407	46.3
	4.50	0.790	0.407	48.4
	6.00	0.769	0.451	41.3
	8.00	0.822	0.504	38.7
	10.0	0.990	0.591	40.3

Table A2.3: Measured and predicted bubble rise velocity data

Appendix B1

Gas Holdup Results



Figure B1.1: Gas holdup versus superficial gas velocity in water



Figure B1.2: Gas holdup versus superficial gas velocity in 0.1 g/l saltwater


Figure B1.3: Gas holdup versus superficial gas velocity in 0.5 g/l saltwater



Figure B1.4: Gas holdup versus superficial gas velocity in 1 g/l saltwater



Figure B1.5: Gas holdup versus superficial gas velocity in 0.5% hardwood pulp



Figure B1.6: Gas holdup versus superficial gas velocity in 1% hardwood pulp



Figure B1.7: Gas holdup versus superficial gas velocity in 2% hardwood pulp



Figure B1.8: Gas holdup versus superficial gas velocity in 3% hardwood pulp



Figure B1.9: Gas holdup versus superficial gas velocity in 4% hardwood pulp



Figure B1.10: Gas holdup versus superficial gas velocity in 6% hardwood pulp



Figure B1.11: Gas holdup versus superficial gas velocity in 9% hardwood pulp



Figure B1.12: Gas holdup versus superficial gas velocity in 0.8% softwood pulp



Figure B1.13: Gas holdup versus superficial gas velocity in 1% softwood pulp



Figure B1.14: Gas holdup versus superficial gas velocity in 2% softwood pulp



Figure B1.15: Gas holdup versus superficial gas velocity in 3% softwood pulp



Figure B1.16: Gas holdup versus superficial gas velocity in 4% softwood pulp

Appendix B2



Gas Holdup Axial Distribution Results

Figure B2.1: Gas holdup axial distribution for water



Figure B2.2: Gas holdup axial distribution for 0.1 g/l saltwater



Figure B2.3: Gas holdup axial distribution for 0.5 g/l saltwater



Figure B2.4: Gas holdup axial distribution for 1 g/l saltwater



Figure B2.5: Gas holdup axial distribution for 0.5% hardwood pulp



Figure B2.6: Gas holdup axial distribution for 1% hardwood pulp



Figure B2.7: Gas holdup axial distribution for 2% hardwood pulp



Figure B2.8: Gas holdup axial distribution for 3% hardwood pulp



Figure B2.9: Gas holdup axial distribution for 4% hardwood pulp



Figure B2.10: Gas holdup axial distribution for 6% hardwood pulp



Figure B2.11: Gas holdup axial distribution for 9% hardwood pulp



Figure B2.12: Gas holdup axial distribution for 0.8% softwood pulp



Figure B2.13: Gas holdup axial distribution for 1% softwood pulp



Figure B2.14: Gas holdup axial distribution for 2% softwood pulp



Figure B2.15: Gas holdup axial distribution for 3% softwood pulp



Figure B2.16: Gas holdup axial distribution for 4% softwood pulp

Appendix B3



Gas Holdup Radial Distribution Results

Figure B3.1: Gas holdup radial distribution for water at $U_g = 0.00272$ m/s



Figure B3.2: Gas holdup radial distribution for water at $U_g = 0.0068$ m/s



Figure B3.3: Gas holdup radial distribution for water at $U_g = 0.0136$ m/s



Figure B3.4: Gas holdup radial distribution for water at $U_g = 0.0204$ m/s



Figure B3.5: Gas holdup radial distribution for water at $U_g = 0.0272$ m/s



Figure B3.6: Gas hold up radial distribution for 0.1 g/l saltwater at $U_{\rm g}$ = 0.00272 m/s



Figure B3.7: Gas holdup radial distribution for 0.1 g/l saltwater at $U_g = 0.0068$ m/s



Figure B3.8: Gas holdup radial distribution for 0.1 g/l saltwater at $U_g = 0.0136$ m/s



Figure B3.9: Gas holdup radial distribution for 0.1 g/l saltwater at $U_g = 0.0204$ m/s



Figure B3.10: Gas holdup radial distribution for 0.1 g/l saltwater at $U_g = 0.0272$ m/s



Figure B3.11: Gas holdup radial distribution for 0.5 g/l saltwater at $U_g = 0.00272$ m/s



Figure B3.12: Gas hold up radial distribution for 0.5 g/l saltwater at $U_{\rm g}$ = 0.0068 m/s



Figure B3.13: Gas holdup radial distribution for 0.5 g/l saltwater at $U_g = 0.0136$ m/s



Figure B3.14: Gas holdup radial distribution for 0.5 g/l saltwater at $U_g = 0.0204$ m/s



Figure B3.15: Gas holdup radial distribution for 0.5 g/l saltwater at $U_g = 0.0272$ m/s



Figure B3.16: Gas holdup radial distribution for 1 g/l saltwater at $U_g = 0.00272$ m/s



Figure B3.17: Gas holdup radial distribution for 1 g/l saltwater at $U_g = 0.0068$ m/s



Figure B3.18: Gas hold up radial distribution for 1 g/l saltwater at U_g = 0.0136 m/s



Figure B3.19: Gas holdup radial distribution for 1 g/l saltwater at $U_g = 0.0204$ m/s



Figure B3.20: Gas holdup radial distribution for 1 g/l saltwater at $U_g = 0.0272$ m/s



Figure B3.21: Gas holdup radial distribution for 0.5% hardwood at $U_g = 0.00272$ m/s



Figure B3.22: Gas holdup radial distribution for 0.5% hardwood at $U_g = 0.0068$ m/s



Figure B3.23: Gas holdup radial distribution for 0.5% hardwood at $U_g = 0.0136$ m/s



Figure B3.24: Gas holdup radial distribution for 0.5% hardwood at $U_g = 0.0204$ m/s



Figure B3.25: Gas holdup radial distribution for 0.5% hardwood at $U_g = 0.0272$ m/s



Figure B3.26: Gas holdup radial distribution for 1% hardwood at $U_g = 0.00272$ m/s



Figure B3.27: Gas hold up radial distribution for 1% hardwood at U_g = 0.0068 m/s



Figure B3.28: Gas holdup radial distribution for 1% hardwood at $U_g = 0.0136$ m/s



Figure B3.29: Gas holdup radial distribution for 1% hardwood at $U_g = 0.0204$ m/s



Figure B3.30: Gas holdup radial distribution for 1% hardwood at $U_g = 0.0272$ m/s



Figure B3.31: Gas hold up radial distribution for 2% hardwood at U_g = 0.0068 m/s



Figure B3.32: Gas holdup radial distribution for 2% hardwood at $U_g = 0.0136$ m/s



Figure B3.33: Gas hold up radial distribution for 2% hardwood at U_g = 0.0204 m/s



Figure B3.34: Gas holdup radial distribution for 2% hardwood at $U_g = 0.0272$ m/s



Figure B3.35: Gas hold up radial distribution for 3% hardwood at U_g = 0.0068 m/s



Figure B3.36: Gas holdup radial distribution for 3% hardwood at $U_g = 0.0136$ m/s



Figure B3.37: Gas hold up radial distribution for 3% hardwood at U_g = 0.0204 m/s



Figure B3.38: Gas hold up radial distribution for 3% hardwood at U_g = 0.0272 m/s



Figure B3.39: Gas hold up radial distribution for 4% hardwood at U_g = 0.0068 m/s



Figure B3.40: Gas holdup radial distribution for 4% hardwood at $U_g = 0.0136$ m/s



Figure B3.41: Gas hold up radial distribution for 4% hardwood at U_g = 0.0204 m/s



Figure B3.42: Gas holdup radial distribution for 4% hardwood at $U_g = 0.0272$ m/s


Figure B3.43: Gas hold up radial distribution for 6% hardwood at U_g = 0.0068 m/s



Figure B3.44: Gas holdup radial distribution for 6% hardwood at $U_g = 0.0136$ m/s



Figure B3.45: Gas hold up radial distribution for 6% hardwood at U_g = 0.0204 m/s



Figure B3.46: Gas hold up radial distribution for 6% hardwood at U_g = 0.0272 m/s



Figure B3.47: Gas holdup radial distribution for 9% hardwood at $U_g = 0.0068$ m/s



Figure B3.48: Gas holdup radial distribution for 9% hardwood at $U_g = 0.0136$ m/s



Figure B3.49: Gas holdup radial distribution for 9% hardwood at U_g = 0.0204 m/s



Figure B3.50: Gas holdup radial distribution for 9% hardwood at $U_g = 0.0272$ m/s



Figure B3.51: Gas holdup radial distribution for 0.8% softwood at $U_g = 0.00272$ m/s



Figure B3.52: Gas holdup radial distribution for 0.8% softwood at $U_g = 0.0136$ m/s



Figure B3.53: Gas holdup radial distribution for 0.8% softwood at $U_g = 0.0272$ m/s



Figure B3.54: Gas holdup radial distribution for 1% softwood at $U_g = 0.00272$ m/s



Figure B3.55: Gas hold up radial distribution for 1% softwood at U_g = 0.0068 m/s



Figure B3.56: Gas hold up radial distribution for 1% softwood at U_g = 0.0136 m/s



Figure B3.57: Gas hold up radial distribution for 1% softwood at $U_g = 0.0204$ m/s



Figure B3.58: Gas hold up radial distribution for 1% softwood at U_g = 0.0272 m/s



Figure B3.59: Gas hold up radial distribution for 2% softwood at U_g = 0.0068 m/s



Figure B3.60: Gas hold up radial distribution for 2% softwood at $U_{\rm g}$ = 0.0136 m/s



Figure B3.61: Gas hold up radial distribution for 2% softwood at $U_{\rm g}$ = 0.0204 m/s



Figure B3.62: Gas hold up radial distribution for 2% softwood at $U_{\rm g}$ = 0.0272 m/s



Figure B3.63: Gas hold up radial distribution for 3% softwood at U_g = 0.0068 m/s



Figure B3.64: Gas hold up radial distribution for 3% softwood at U_g = 0.0136 m/s



Figure B3.65: Gas hold up radial distribution for 3% softwood at U_g = 0.0204 m/s



Figure B3.66: Gas hold up radial distribution for 3% softwood at U_g = 0.0272 m/s



Figure B3.67: Gas hold up radial distribution for 4% softwood at U_g = 0.0068 m/s



Figure B3.68: Gas hold up radial distribution for 4% softwood at U_g = 0.0136 m/s



Figure B3.69: Gas hold up radial distribution for 4% softwood at U_g = 0.0204 m/s



Figure B3.70: Gas hold up radial distribution for 4% softwood at $U_{\rm g}$ = 0.0272 m/s