# ANALYSIS OF EJECTOR-STYLE MICROBUBBLE GENERATORS: MASS-TRANSFER PROPERTIES, MATHEMATICAL MODELING, AND DESIGN ALGORITHM

By

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### ABSTRACT

Gas-to-liquid mass transfer is a rate-limiting step for many commercial-scale operations in the chemical, biochemical, pharmaceutical, and wastewater-treatment industries. The use of microbubbles with a diameter on the100 µm scale has been shown to provide high volumetric mass-transfer rates due to its high gas contact area per volume. However, the use of microbubbles in commercial processes has been hampered by the lack of design algorithms with which to fabricate high-performance, microbubble-sparged gas-liquid contacting equipment. The goals of this study were to identify the type of microbubble generator best suited to provide high volumetric mass transfer rates in commercial-scale equipment, characterize the mass-transfer properties, develop models able to predict the mass-transfer rate as a function of the key independent variables, and use the models to develop a design algorithm suitable to use microbubble sparging in industrial processes.

The study began with a literature review of microbubble generators that considered factors including the mechanism, safety, cost, and scalability, with the goal of identifying generators suited to cost-effectively provide extremely high mass transfer in commercial-scale equipment. Microbubble generators that used liquid turbulence were found to have the best combination of properties for such applications.

In collaboration with the Michigan Biotechnology Institute, a 300-L bioreactor was customized for use with either a RiverForest microbubble ejector and a conventional ring sparger. *E.coli* batch growth experiments were conducted to compare the growth rates using the two aeration methods. The *E.coli* growth rate observed during microbubble aeration was about twice that observed with the traditional ring sparger.

Mathematical models describing the performance properties of both a microbubble ejector and a Modified Jameson Cell were developed. The models included energy requirements, mass transfer rates, gas and liquid flow patterns, and clearance of spent bubbles. The models predicted that the ejector would be more energy-efficient for applications requiring higher mass-transfer rates and lower gas volume fractions, whereas the Modified Jameson Cell would be more energy-efficient for applications requiring lower mass-transfer rates and higher gas void fractions.

A novel flow system was developed to measure the mass-transfer rate of microbubble produced by an ejector generator. A mathematical model was developed to reproduce experimental trends and estimate the effective microbubble diameter generated as a function of the gas and liquid flow rates. New axial mixing and two-phase friction factor correlations were developed for the model fidelity. The results were used to develop a correlation to predict the effective microbubble size as a function of system properties. The predictive power of this correlation has utility for industrial process design and scale-up applications.

The friction factor and microbubble diameter correlations developed in this study were used to develop additional models to simulate the microbubble mass-transfer in large reactors that are sparged with arrays of microbubble ejectors. Collectively, the models developed in this study provide powerful new design tools that enable rational design, optimization, and scale-up of ejector microbubble sparger arrays for commercial-scale reactors that require high mass-transfer rates.

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# LIST OF SYMBOLS

A: absorbance

a: interfacial area per unit reactor volume

 $a_{pipe}$ : cross-sectional area of a pipe

C: bulk concentration of the transferred gas dissolved in the liquid

 $C^*$ : equilibrium solubilities

 $C_0$ : constants correlated to the perforated plate geometry

 $C_C$ : constants correlated to the perforated plate geometry

Cero: pipe erosion velocity constant

 $C_f$ : Chen's arbitrary constants

 $c_{dye}$ : dye concentration

D: pipe diameter

*D*<sub>bub</sub>: bubble diameter

 $\tilde{D}_{b,m}$ : dimensionless bubble diameter inside the MJC

*D<sub>pore</sub>*: pore diameter

 $d_{max}$ : maximum stable bubble diameter

 $d_{noz}$ : ejector nozzle diameter

*E*<sub>diss</sub>: dissipation energy

*E*<sub>surf</sub>: surface free energy per unit area,

Eu: pressure loss coefficient

Frg: Froude Number *f*: friction factor *f<sub>fan</sub>*: Fanning friction factor g: gravitational constant *g<sub>c</sub>*: conversion factor  $\tilde{H}$ : dimensionless height inside the column H<sub>pump</sub>: pump head *H*<sub>tank</sub>: vessel height *h*: axial position inside jet.  $h_{f}$ : total fitting head-loss *h<sub>jet</sub>*: jet height value  $h_{pipe}$ : friction energy head loss  $k_{fit}$ : fitting loss coefficient  $k_l$ : liquid-film mass transfer coefficient *L*: ejector length *L*<sub>tube</sub>: tube length *l*: light path distance *N*<sub>bub</sub>: total number of bubbles *m*: slope  $m_g$ : gas molar flow rate

*n<sub>f</sub>*: Chen's arbitrary constants

OUR: Oxygen Uptake Rate

*P*: Power consumption

*P<sub>atm</sub>*: atmospheric pressure

P<sub>fri</sub>: friction induced pressure drop

*P<sub>gauge</sub>*: gauge pressure

*P<sub>hyd</sub>*: hydrostatic pressure

*Plap*: Laplace pressure

*P<sub>pump</sub>*: pump power consumption

 $P_{total}$ : total pressure

 $\Delta p$ : pressure drop

 $\Delta p_m$ : porous plate pressure drop

 $\Delta p_{mix}$ : two-phase pressure drop

Pe: Peclet number

Q: volumetric flow rate

 $Q_{read}$ : rotameter flow rate reading

Re: Reynolds number

*r*: radial position inside the jet

*r*<sub>bub</sub>: bubble radius

*t*<sub>pore</sub>: pore depth

*t<sub>res</sub>*: residence time of the stagnant fluid inside the void zone *u*: superficial velocity *u*<sub>ero</sub>: pipe erosion velocity *u<sub>jet</sub>*: jet velocity at any point inside a jet  $u_{max}$ : maximum liquid velocity at the jet centerline *V*: reactor volume Vall: total volume of the mass-transfer unit cell *V<sub>cone</sub>*: jet cones volume  $V_{ejector}$ : ejector volume V<sub>max</sub>: maximum theoretical reactor volume  $V_{oid}$ : volume between the jet cones  $\tilde{V}$ : dimensionless superficial velocity *Vana*: van der Waals constant *Van<sub>b</sub>*: van der Waals constant *W*: power consumption *We*: Weber number *We<sub>c</sub>*: Critical Weber Number *We<sub>jet</sub>*: Jet Weber Number X: cell concentration *X*<sub>o</sub>: percentage of gas being reacted

 $y_{O2}$ : O<sub>2</sub> content inside the gas

 $\varepsilon$ : volume fraction

 $\rho$ : density

 $\beta$ : square root of the quotient between pore area and plate area

 $\phi$ : energy dissipation rate per volume

 $\eta$ : pump efficiency

 $\sigma$ : surface tension

 $\mu$ : viscosity

 $\mu_{net}$ : cell net growth rate

 $\varepsilon_{att}$ : molar attunement coefficient

subscript:

*Big*: Lab-made microbubble generator (10 mm)

*bottom*: bottom of a large reactor

g: gas

*i*: gas species

*l*: liquid

mix: gas-liquid mixture

*r*: radial direction of the flow

*River*: Riverforest microbubble generator (7 mm)

Small: Lab-made microbubble generator (7 mm)

*top*: top of a large reactor

*z*: axial direction of the flow

 $\theta$ : angular direction of the flow

# LIST OF ABBREVIATIONS

CPLJ: Confined plunging liquid jet

DOP: dissolved oxygen profile

DOS: dissolved oxygen sensor

FOAK: First-Of-A-Kind

HUSH: hollow cylindrical ultrasonic horn

ISR: industrial-scale reactor

MBE: microbubble ejector

MBG: microbubble generator

MJC: modified Jameson cell

PFR: Plug Flow Reactor

RO: Reverse-osmosis

RTD: Residence time distribution

SDS: sodium dodecyl sulfate

VDC: Vessel Dispersion Coefficient

VFD: variable frequency drive

VVM: volume of air per volume of reactor per time

# **CHAPTER ONE: LITERATURE REVIEW**

#### **1.1 Background Overview**

Many commercially important chemical and biochemical reactions that are carried out in a continuous liquid phase consume sparingly soluble gaseous reactants (e.g., O<sub>2</sub>). The rate of gasto-liquid mass transfer is frequently the rate-limiting step when these reactions are conducted in commercial-scale reactors. An effective process-intensification strategy for such reactions is to accelerate the mass-transfer step by dispersing the gas phase as microbubbles on the order of 100 µm in diameter. However, industrial implementation of this strategy has been hampered by the lack of a published design algorithm for commercial-scale microbubble-sparged reactors. One goal of this dissertation is to assess the suitability of alternative microbubble-generation methods for cost-effectively achieving high volumetric mass transfer rates ( $k_{la} > 1000 \text{ h}^{-1}$ ) in commercial-scale reactors. Emphasis is placed on methods that use liquid turbulence to generate microbubble dispersions, such as ejector and modified Jameson cell generators [1]. Published engineering correlations are used to predict and compare performance metrics of these systems, including volumetric mass-transfer rates, power-to-volume ratios, and suitability for scale-up.

### 1.1.1 Current Need for Rapid Gas-liquid Transfer

Many multiphase chemical and biochemical reactions involve gaseous reactants that have low solubility in a continuous liquid phase. Examples include fermentation reactions that consume O<sub>2</sub> and/or CH<sub>4</sub> carried out in aqueous solutions under mild temperatures and pressures. The

equilibrium solubilities ( $C^*$ ) of O<sub>2</sub> and CH<sub>4</sub> at 35°C and one atmosphere pressure are 33 mg/L (1.1 mM) and 17 mg/L (1.0 mM), respectively.[2] A recent DOE ARPE-E research program funded efforts to develop economically competitive fermentations that achieved an extremely high volumetric mass transfer rate ( $\dot{Q}$ ) for CH<sub>4</sub> of  $\dot{Q}_{CH4}$ >50 g<sub>CH4</sub>/(L<sub>reactor</sub> h) [3]. The volumetric mass transfer coefficient ( $k_{l}a$ ) needed to achieve this  $Q_{CH4}$  value can be estimated using the design equation for interphase gas mass transfer,  $\dot{Q} = k_{l}a(C^* - C_L)$ , where  $k_l$  is the liquid-film mass transfer coefficient, a is the interfacial area per unit reactor volume,  $C_L$  is the bulk concentration of the transferred gas dissolved in the liquid, and ( $C^* - C_L$ ) is the driving force for mass transfer. When gas mass transfer is rate-limiting,  $C_L \approx 0$ , so  $Q \approx k_l a C^*$ . Using this equation, the abovementioned ARPA-E technical target translates into a  $k_l a$  value of 2,900 h<sup>-1</sup>[3].

# 1.1.2 Industrial Practices and Challenges on Gas-intense Operations

The literature indicates that achieving  $k_l a$  values greater than 1000 h<sup>-1</sup> cost-effectively is challenging. Commercial-scale aerobic fermentations are typically O<sub>2</sub>-mass-transfer limited, [4, 5] and stirred-tank reactors are generally used in such cases, because they achieve significantly higher  $k_l a$  values than columnar bubble columns or airlift reactors [4]. The intense turbulence generated by a stirred tank's impeller breaks large bubbles into smaller ones, thereby decreasing the average bubble diameter ( $D_{bub}$ ). Because a is given by Equation 1-1, where  $\varepsilon$  is the gas volume fraction, for a given  $\varepsilon$  value, decreasing  $D_{bub}$  by a given percentage increases a, and thus  $k_l a$ , by the same percentage.

$$a = \frac{6\varepsilon}{D_{bub}} \tag{1-1}$$

However, using stirred tanks to achieve extremely high  $k_la$  values scales up unfavorably. For geometrically similar tanks, power consumption (*P*) increases with impeller diameter to the fifth power, whereas reactor volume, *V*, only increases with impeller  $D_{bub}$  to the third power [6]. Moreover,  $k_la$  has a relatively weak dependence on *P*/*V* (e.g., the square root of *P*/*V*) [5]. As a result, the power costs required to generate high  $k_la$  values in large, stirred-tank reactors can be prohibitively high.

Typical ranges of *P/V* values used in commercial scale reactors are reflected in literature reviews of gas-liquid mass transfer in stirred vessels. Van't Riet's review presented results over a *P/V* range of 0.4 to 10 kW/m<sup>3</sup>, which corresponded to  $k_ia$  values over a range of 72 h<sup>-1</sup> to 2200 h<sup>-1</sup> [5]. Garcia-Ochoa's review correlated data in seven studies over a *P/V* range of 0.2 to 1.1 kW/m<sup>3</sup>, which corresponded to a  $k_ia$  range of 22 to 150 h<sup>-1</sup> [4]. Both of these  $k_ia$  ranges are well below the primary technical target value (2900 h<sup>-1</sup>) specified in the ARPA-E program, [3] suggesting that the conventional stirred tanks are not well suited to achieve the target  $k_ia$  value cost-effectively in commercial-scale reactors.

#### 1.1.3 Microbubble Advantages on Gas-liquid Mass Transfer

Microbubble dispersions offer several advantages over conventional bubbles for intensifying a wide range of industrial processes.[7, 8] Microbubbles' extremely small  $D_{bub}$  (on the order of 100 µm) imparts several desirable properties, including extremely high large *a* and  $k_{la}$  values (Equation 1-1). At such small  $D_{bub}$  values, the intra-bubble pressure exceeds the pressure of the surrounding liquid by an amount that is inversely proportional to  $D_{bub}$ , as described by the Young-Laplace ( $p_{lap}$ ) equation, where  $r_{bub}$  is bubble radius and  $\sigma$  is surface tension.

$$\Delta p_{lap} = \frac{2\sigma}{r_{bub}^2} \tag{1-2}$$

This "Laplace pressure" accelerates a microbubble's  $C^*$ ,  $(C^* - C_L)$ , mass transfer rate, and rate of shrinkage as gas is transferred out of the microbubble. Moreover, microbubble shrinkage culminates in a violent bubble collapse that generates localized thermal gradients and free radicals[9, 10] that can be used to accelerate some chemical reactions, to accelerate biomass pretreatment[11], and to accelerate degradation of toxic compounds in wastewater treatment.[12]

Microbubble sparging has demonstrated great potential in gas-intense operations where gas mass transfer is usually the rate limiting step [13, 14]. Microbubble spaging has been to used intensify a variety of processes, including biomass pretreatment[11], degradation of toxic compounds in wastewater,[12] biological water treatment [9, 15-17], fish farming [18, 19], nicotine biosynthesis [20], ocean vessel design[21], and various bioreactor processes [7, 22-25].

# **1.2 Microbubble Generation Techniques and Technical Benchmarks**

Alternative methods to generate microbubbles for specific applications have been reviewed [26-29]. Parmar and Majumder described alternative microbubble-generation methods for waste treatment applications, but their study did not estimate power requirements or cost efficiency of the alternative methods for generating a target  $k_{la}$  value [26]. Zimmerman et al. (2008) introduced

multiple innovative ways to produce microbubbles at a relatively low rate, but this article did not discuss the feasibility of achieving extremely high  $k_{la}$  values in commercial-scale reactors or power requirements [27]. Terasaka et al. (2011) reviewed multiple microbubble-generation methods and conducted experiments to measure the O<sub>2</sub> transfer rate at a given power consumption level, but their reactor column was only 0.056 m<sup>3</sup>, and the feasibility of scaling the system up was not considered [28]. Burns et al. reviewed some microbubble-generation methods based on electrolysis and rapid depressurization of liquid that was supersaturated with a gas. However, the methods Burns et al. discussed are utilized in separations instead of large-scale, gas-intense reactor processes [30]. The energy requirement from electrolysis-based microbubble-generation methods could also be too high for mass transfer processes. Rodriguez-Rodriguez et al. discussed various microbubble-generation methods for small-scale medicine productions as well [29]. Ansari et al. investigated microbubble-generation system that used individual tubes having an inner diameter on the order of 100 µm that were individually cut using a laser. This microfluidic fabrication approach would not be sufficiently scalable for commercial reactors [31].

The abovementioned studies have not satisfactorily addressed the important question of which microbubble-generation method(s) is best suited for commercial-scale chemical or biochemical reactors that require sparingly soluble gases to be transferred cost-effectively at extremely high  $k_la$  values. The goal of this study is to address that question, which will contribute toward the ultimate objective of developing the first algorithm for design and scale-up of microbubble-sparged reactors that can meet ambitious to  $k_la$  targets (>1000 h<sup>-1</sup>) cost-effectively at the commercial scale.

# 1.2.1 Bubble Size and Interfacial Area

As described above, the average  $D_{bub}$  and resulting *a* values are primary performance metrics. However, because the power required to generate the surface area commonly represents a major component of the operating costs of reactors requiring high  $k_{la}$  values, the ratio of power consumed to interfacial area generated is also extremely important. In addition to the average  $D_{bub}$ , the breadth of the  $D_{bub}$  distribution can also significantly affect system performance. Previously, twophase flow microbubble-generation methods have been criticized for having a broad  $D_{bub}$ distribution[32], which would make it difficult to measure the  $D_{bub}$  via experiments.

# 1.2.2 Bubble Rising Velocity

Bubble terminal rise velocity is an important factor for microbubble mass transfer because it governs gas-liquid contact time [33]. Bubble rise velocity is correlated to drag force and buoyancy force, which are further related to bubble size [34]. For rising bubbles whose Reynolds number (*Re*) is less than about 1.5 the rise velocity is reasonably represented by Stokes' Law [35], which indicates that the rise velocity of bubbles varies inversely with the square of the bubble diameter [36]. For an aqueous solution at about 25°C, this *Re* regime has an upper bound of about 130  $\mu$ m. This correlation means sparging smaller bubbles would drastically improve the residence time for two phases to react and reduce material waste, thus improving the cost-efficiency of the two-phase operations [15].

### 1.2.3 Bubble Coalescence

Bubble coalescence can substantially reduce the volumetric mass transfer rate in a microbubble-sparged reactor through two mechanisms. First, it results in a larger average  $D_{bub}$ value, and consequently, as shown in Eq. 1-1, smaller a and  $k_{la}$  values. Second, by increasing  $D_{bub}$ , coalescence reduces the intra-bubble pressure, and consequently, the  $C^*$  value and mass-transfer driving force. The rate of coalescence depends on two factors: the rate of bubble collisions and the fraction of bubble collisions that result in a coalescence event rather than an elastic collision. When two bubbles collide, if the thin liquid film separating them drains to a critical value, the liquid film will rupture, and the two bubbles will merge [37, 38]. Bubble coalescence mechanisms and dynamics have been investigated using X-ray imaging [39], the lattice Boltzmann method [40], and the population balance method [41]. Prince & Blanch studied the impact of bubble coalescence phenomena on bubble-column performance and developed model for microbubble coalescence and break-up rate [42]. The rate of bubble coalescence observed in pure water can be reduced substantially by adding a sufficient amount of electrolytes, whose charges result in electrostatic repulsion forces between adjacent bubbles that prevent intra-bubble liquid films from rupturing, thereby inhibiting coalescence [43, 44]. In a previous study conducted by Zahradnik et al., it was found that adding small amount of electrolyte to xanthan-alcohol system could reduce bubble coalescence to almost non-exist [45]. Surfactant is another method to stabilize the bubble sizes and prevents bubble coalescence[46]. The characteristics of surfactant-stabilized bubbles were investigated before by numerous research groups previously as well [47-50].

#### 1.2.4 Microbubble Generator Power Consumption

Implementing microbubble generator (MBG) could also reduce operation and capital cost as well. Previous study conducted on bubble sparging column indicated that the liquid inside the system is well mixed, thus no further mixing apparatus is required inside a microbubble sparging column [51]. This finding indicates microbubble generation could replace impeller rotation as the method of improving gas-liquid contact. The installation cost, along the operating cost of impellers could be removed from the overall operation and capital cost.

### **1.3 Microbubble Generation Mechanisms**

Many microbubble-generation methods have been developed over the years based on different fluid dynamics principles. In this study, we focus on the microbubble-generation methods based on fluid turbulence due to their simplicity and cost-efficiency when scaling up. This study also includes a collection of other microbubble generation based on other principles but were left out of cost efficiency discussion due to limit in the implementation of these microbubble-generation systems on scale-up systems.

### 1.3.1 Turbulent Energy Based

Turbulence energy types of microbubble-generation methods rely on turbulence energy of the fluids to break up two-phase fluids down into microbubbles. These types of microbubblegeneration systems usually have gas and liquid flow through confined spaces at high liquid superficial velocity in order to maintain a level of turbulence and shred larger air pockets inside the liquid into microbubbles. The efficiency of this process is dependent on the equipment dimensions and fluid flow rate, which will affect the bubble size and energy cost of the systems. The effect of nozzle shape and size on bubble characteristic has been previously investigated [52], providing a good basis for optimization and design.

### 1.3.1.1 Confined Plunging Liquid Jet and Modified Jameson Cell

Confined plunging liquid jet (CPLJ) is a microbubble-generation method that utilizes shear and turbulence to create microbubbles inside a flotation cell [53]. Inside a conventional plunging jet bubble generator, liquid and gas flow downwards into a cell and bubbles would form due to complex hydrodynamics [54]. Over the years, researchers have developed industrial production units based on CPLJ such as Jameson Cell [55] and cyclo-microbubble Flotation Column [56]. Majumder et al. has measured the bubble size and gas-liquid interfacial area inside a down flow bubble column [57]. This microbubble-generation method has had scale up application such as mineral flotations [58] and ozone water purification processes [59, 60] due to its improvement on gas mass transfer. However, the bubble generated using conventional plunging jet would sometimes still generate bubbles on millimeter scale, which is too large compared to other microbubble-generation methods [57, 59].

For all plunging jet columns, the penetrating depth of gas should also be considered when designing the column. Since there is only one gas source from above, the total amount of gas

transferred into the column would deplete after a certain depth. After this depth, it is pointless to add in additional height to the column since the rest will not receive any aeration at all. The calculated depth can be expressed in the following form, where  $u_g$  is gas superficial velocity across the column and  $\rho_g$  is the gas density:

$$depth_{MJC} = \frac{u_g \rho_g}{k_l a C^*} \tag{1-3}$$

A modified Jameson cell (MJC) system described by Li et al. includes a column through which liquid flows in a downward direction and bubbles of down-flowing liquid reactant liquid through which bubbles of the reactant gas rise that combines features of a Jameson Cell and conventional bubble sparger [1]. The MJC system consisted of a liquid column used a conventional porous bubble sparger at the bottom of the cell, and a perforated plate for liquid jet generation purpose at the top. Coarse bubbles on millimeter scale would generate by the porous sparger and then float to the top of the cell due to buoyance forces. These coarse bubbles would then generate a layer of foams on top of tank column liquid. The liquid jet from above would breakdown the foam layer into microbubbles and the current would carry the microbubble downwards for gas transfer. The schematic can be seen in Figure 1-1. This MJC column design has already been utilized in a pilotscale bioreactor according the patent US20140212937. According to the patent, the MJC column managed to produce microbubbles on average diameter of 60 µm while Hernandez-Alvarado et al. managed to produce microbubble with average diameter of 600 µm. Sodium dodecyl sulfate (SDS) was added as a surfactant to stabilize the bubble formation in both the patent (100 ppm) [1] and

the bubble size estimation study (10 ppm) [61] for the MJC. This adds in additional material and separation cost for the production and could also reduce the overall gas mass transfer rate [8]. The patent contained more SDS and managed to produce smaller bubbles.



Figure 1-1. Simplified schematics of a MJC Microbubble Generator

#### 1.3.1.2 Spiral Ejector

For swirl ejector microbubble generators, the fundamentals behind microbubble formation vary from type to type. An earlier design of swirl microbubble ejector was used for wastewater treatment [28]. The main body of the swirl ejector is a swirling chamber. The gas and liquid streams were injected into a swirl chamber perpendicular to each other, and the gas-liquid mixture is broken down into microbubbles due to the centrifugal force acting on the liquid flow [62]. The liquid usually enters the chamber from the side while the gas enters from the bottom. The cylindrical liquid flow inside a tube has been thoroughly investigated before via DNS simulations and numerical simulations [63-65]. A more recent invention by Hato claimed that by isolating the gas and liquid injection flow, the microbubble ejector could generate bubbles on nano-scales. This is contributed to the conservation of kinetic energy [66]. Swirl microbubble ejector has wide utilizations in dissolved air flotation operation such as water treatment [67, 68]. The same type of microbubble generator has also had applications in Wafer cleaning devices [69]. Another usage for spiral microbubble ejector is for underground waste water treatment [15]. The aforementioned water treatment system managed to produce microbubbles of diameters from 0.5 to 100  $\mu$ m. Another investigation from Levitsky et al. managed to produce microbubbles on scale of 20  $\mu$ m and the research group managed to obtain the bubble size distribution correlations with operating conditions [70].

Figure 1-2 demonstrates a simplified version of spiral microbubble ejector, where liquid is introduced on the top of the chamber while gas is injected from the bottoms.



Figure 1-2. Simplified schematics of a spiral ejector style microbubble generator

Terasaka et al. conducted an investigation on water sludge treatment and water aeration rate and they have compared the effect of microbubble sparging methods on O<sub>2</sub> transfer rate. The study concluded that spiral ejector style microbubble-generation method can improve O<sub>2</sub> transfer rate better than other methods, albeit at higher energy cost[28]. Another paper by Li et al. on this water treatment method has managed to find the bubble size distribution under different combination of aeration methods and fluid components [71].

#### 1.3.1.3 Venturi

A Venturi type microbubble generator consists of a hollow tube with a converging-diverging neck. This type of microbubble-generation techniques utilizes the cavities of the Venturi neck to break up the bubbles to a desirable size [72]. The liquid flow, upon hitting the narrowed neck, would increase in pressure and create a pressure wall due to Bernoulli effect. The  $u_l$  can reach up to sonic speed at the entrance of the converging-diverging neck, which is not found in ejector MBG [72]. The gas mixed in would bounce off the pressure wall and create microbubbles due to shockwave [9]. The bubble size prediction based on fluid flow and instrument dimensions was previously obtained through image analysis [73]. A more recent study by Gordiychuk et al. has correlated the bubble size distribution with dimensionless operation parameters such as  $\varepsilon$  and Reusing image analysis technique as well [74]. The Venturi system used managed to produce microbubbles on scale of 100 to 400 µm. The utilization of Venturi microbubble generator has shown improvement in yeast fermentation [23]. Figure 1-3 shows a simplified demonstration of Venturi type microbubble generator, which comprises of a two-phase fluid inlets and a converging-diverging nozzle [26].



Figure 1-3. Simplified schematics of a Venturi style microbubble generator

Patent US20160325242A1 indicated that Venturi type microbubble generator has applications in fields of furniture and decorations, such as bathtubs and swimming pools [75]. The bubbles produced using this method is on scale of micrometer scale, which makes it less attractive than other bubble generation methods due to its larger bubbles [72].

The bubble movement, velocity, deformation, and collapse through the converge-diverge nozzle have been investigated through image analysis previously [76]. It was found out that decreasing  $Q_l$  would increase bubble collapse, thus reducing the  $D_{bub}$ . This finding suggests a trade-off between production rate (correlated to  $Q_l$ ) and  $D_{bub}$ , which makes it difficult to optimize. Another problem with Venturi tube is the cavitation of bubbles inside the nozzle [72]. It would be counter-productive to use Venturi tubes to generate bubbles since the goal of utilizing microbubble

is to increase gas dissolution inside the liquid. If cavitation removes dissolved gas inside liquid due to cavitation, using Venturi tube would defeat the whole purpose of microbubble aeration.

#### 1.3.1.4 Spinning Disk

Sebba et al demonstrated the setup of a spinning disk gate microbubble generator in US5314644 [77]. Spinning disk gate microbubble generator utilizes shear momentum of liquid to disperse pocket of gas through porous materials. In this particular design, a motor is installed at the bottom of a cylindrical vessel, and a round porous plate is attached to the rotor. Gas-liquid-surfactant mixture was propelled into the rotating cylinder complex and broken down into microbubbles [77].

Previous experiments conducted on rotating disk microbubble generator has found that this bubble generation technique could increase  $O_2$  transfer rate at lower agitation rate [78]. This indicates that implementing a rotating microbubble generator would be more energy conservative compared to mechanical agitating aeration processes. The spinning disk microbubble generator has shown potentials in increasing biomass fermentation [24, 25] and improving hairy root metabolites growth rate [20].

This microbubble generator design, however, is relying on surfactants to stabilize the bubble, which will not be economically viable due to material and separation costs. Additionally, surfactant content inside microbubble flow could reduce the overall O<sub>2</sub> transfer efficiency [15].

#### 1.3.2 Microbubble Generators Based on Other Principles

Microbubble-generation methods based on fluid-dynamics mechanisms other than turbulence have been developed. However, these methods are not considered well suited for large-scale industrial application for reasons described below.

#### 1.3.2.1 Oscillation

A fluidic-oscillator mechanism has been used to produce microububble streams based on surface instability and Coanda effect [79, 80]. A fluidic oscillator employs a fluid amplifier whose geometry triggers a side-to-side mechanical oscillation under the influence of a flow field [81]. The main body of some fluidic oscillators have no moving parts, which proponents claim makes them easy to manufacture, durable and cost-efficient [82, 83]. As a gas stream enters the liquid phase, the oscillations influence bubble formation dynamics, interrupting the bubble development process and creating smaller bubbles than would have resulted in the absence of the oscillation[22]. Zimmerman et al. described use of a fluidic oscillator for aeration operations [84] and its potential application to bioreactors [81] and coal flotation [85]. Al-Mashhadani et al. measured masstransfer properties of oscillation-induced microbubbles in CO<sub>2</sub> mass transfer inside a bubble column [86]. The use of microbubbles produced by fluidic oscillation in the pretreatment of lignin biomass has been explored on the basis that O2 radicals generated by microbubbles could increase reactivity of biomass [11] and biogas [87].

The bubble sizes generated via fluid oscillation have been correlated with the oscillation

frequency, gas flow rate, and oscillator design [88]. Brittle et al. showed that an 'optimal' oscillation frequency for liquid oscillator that results in minimal bubble size microbubbles [79]. However, bubble size was found to fluctuates erratically with minor changes in gas flow rate [89]. This finding suggested that rigorous control of gas velocity through the fluidic oscillators would be required to obtain optimal bubble formation. Moreover, the Conada-effect oscillation occurs at the length scale of individual bubbles, suggesting that scale-up of the method would require the number of precision-fabricated oscillators to increase linearly with gas throughput. Each fluidic oscillator unit presumably requires precision fabrication, and a large number of small fluidic oscillators. Such a scale-up strategy would seem impractical for commercial-scale reactors that operate at very large  $k_ia$  values.

#### 1.3.2.2 Electrolysis

Electrolysis can be used to form bubbles of  $H_2$ at the cathode and  $O_2$  at the anode of an electrolytic cell. The size and density of the bubbles can be controlled by adjusting the electrolyte concentration, power output, and electrode material and local hydrodynamic forces [90, 91]. Microbubbles 20  $\mu$ m in diameter have been produced using electrode having sharp tips [92]. The delivery rate of the bubble is also a function of electrode shape and local hydrodynamic forces. However, this approach is impractical for mass-production of microbubbles for large-scale industrial applications [91] for several reasons. The approach would be useful only in rare

applications in which simultaneous production of a 2:1 molar ratio mixture of  $H_2$  and  $O_2$  gases is needed. The production of significant quantities of this explosive gas mixture would create a safety hazard. Moreover, the manufacturing costs of custom electrodes having sharp tips and electrical costs required to drive the hydrolysis reactions and the cost of the electricity would likely to be prohibitively high.

#### 1.3.2.3 Microfluidic

Microbubbles can be generated in a microfluidic flow system when a continuous liquid stream impinges on a continuous gas stream (e.g., in a T junction), and hydrodynamic forces break up the gas stream into discrete bubbles. Garstecki et al [93] investigated the bubble-formation mechanism in a microfluidic T-junction microbubble generator and developed a scaling law to predict the bubble size as a function of diameter of the liquid channel's diameter and the gas and liquid flow rates. This microbubble-generation method has potential to make relatively small quantities of microbubbles having a uniform bubble diameter. However, the costs to microfabricate and to install large numbers of such microbubble generators and to pump large quantities of liquid through them in laminar flow would be prohibitively high for large-scale reactor applications.

### 1.3.2.4 Ultrasonic

By attaching an ultrasound inducer to a steel capillary, microbubbles can be injected into high surface tension fluids due to shockwave-generated bubble collapsing [94]. The size of the microbubble is controlled by inducer frequency and capillary dimensions. Pulse-induced bubble size prediction and formation from a capillary was investigated using both computational fluid dynamics and image analysis [95]. From photographic evidence obtained by the same research group, it was observed that a single stream of microbubbles is produced when the needle is vibrated at a certain frequency. This evidence indicates the total gas flow rate is very low and cannot sustain industrial scale production [94]. Bubble coalescence under ultrasound field could also be a concern when designing reaction systems [96]. The coalescence phenomena of microbubbles under ultrasound field and bubble breaking model were calculated based on inertial drainage model [38, 97]. Kobayashi et al. also discussed using ultrasonic irradiation to separate microbubbles on industrial scales should the demand rises [98].

An upgrade version of ultrasound microbubble generator was designed by Makuta el al. in order to improve the total gas production rate using cylindrical horn [32]. By using a horn instead of needle to sparge gas and to create ultrasonic vibrations, the total gas production rate is increased. From graphic evidence, the total gas production rate was improved since the gas is coming out of orifice pores whose total area is much larger than a sparging needle [32]. However, the increase in gas flow rate is insufficient according to a later research based on this type of design [99]. This research based on hollow cylindrical ultrasonic horn (HUSH) managed to improve the gas transfer rate and reduce coalescence through adding multiple orifices inside a HUSH [99]. Figure 1-4 demonstrates a simplified schematics for a HUSH horn.



Figure 1-4. Simplified schematics for the experiment setup by Tatsuya et al. [99], depicting the fundamental elements of an ultrasonic microbubble generator.

Achaoul et al. have recently developed a new method to combine the electrolysis and ultrasonic microbubble-generation method [100]. This type of ultrasonic microbubble generator combines electrolysis microbubble-generation method and the above ultrasonic horn method to enhance the control over  $D_{bub}$  and liquid volumetric flow rate (Q). By synchronizing the electrode and ultrasound frequency, the  $D_{bub}$  and Q can be finely controlled [100]. Under such synchronized conditions, the system can produce pure H<sub>2</sub> gas for energy usage purposes, yet it is not economical to invest in electricity in order to produce H<sub>2</sub> gas. However, this type of microbubble-generation method is very energy intensive, thus not viable for industrial purposes. In addition, in order to control the bubble generation with precision, the ultrasound inducer must be finely tuned, which requires a PID controller. The article proposed the electrolysis-ultrasound production type pointed out that this hybrid microbubble-generation method is more suitable for medicine and biology field [100].

# **1.4 Microbubble Ejector**

In a previous study, Shirtum et al. reported that highly turbulent, gas-liquid flow in a confined area, such as an microbubble ejector (MBE), could generate steady flow of bubbles on micron scales [101]. The bubble formation is contributed to the turbulence energy. As fluid turbulent energy exceeds the surface tension energy, the gas-liquid mixture would break up into microbubbles [102].

One of the MBGs utilizing turbulent energy is the MBE. A schematic of MBE can be seen in Figure 1-5 High velocity liquid flows through narrow channel and mix with gas that was transported in by line pressure difference. The gas-liquid mixture is then broken down into microbubbles due to high turbulence and the liquid-bubble mixture is then ejected into the vessel. A typical microbubble ejector contains a suction nozzle, a liquid nozzle, a mixing tube, and a bubble diffuser [103]. The liquid nozzle is connected to the inlet pipe of whatever the system the ejector is aerating. The gas nozzle is connected to gas pipeline. The mixing tube is narrower than the inlet nozzles in order to increase the fluid speed and create turbulence for bubble creations. The exit nozzle would sparge out bubble-liquid mixture into the reactor systems. An MBE has no moving part, thus making it very simple and cheap to maintain [104]. The two-phase flow dynamics and O<sub>2</sub> transfer inside an MBE has been thoroughly studied before, making it much easier to design suitable MBE for different operating conditions [103, 105].



Figure 1-5. Simplified schematics of an MBE microbubble generator

The size of bubbles generated by fluid turbulence is heavily correlated to fluid superficial velocity. According to previous study on fluid characteristics done by Chen et al., the combination of high liquid  $(u_l)$  and low gas  $(u_g)$  superficial velocity would produce microbubbles with the smallest diameter [102].

Kim et al. have studied the gas-hold up, flow regime, and gas suction rate for two-phase flow inside an MBE, and the result could be used for design [105]. The bubbly flow throw narrow channel was investigated through computational fluid dynamics and numerical simulation [106]. Prediction of  $D_{bub}$  from operating conditions, characteristics, and equipment dimensions is feasible with the available knowledge. The pressure drop for two-phase flow has been previously studied through experiments and computational fluid dynamics [107]. The bubble coalescence inside a jet ejector has been investigated before through computational fluid dynamics [41]. Since both bubble size and pressure drop are available for estimations, the cost-efficiency estimation for MBE system scale-up would be feasible. Patent US6017022 claimed that MBE has been utilized in industrial production, such as monomer processing, air stripping, and water sludge treatment [101].

### 1.4.1 Overall Mechanics Behind Microbubble Ejectors

The  $D_{bub}$  obtained in an MBE can be estimated using the correlation developed by Chen et al., which expresses the maximum stable bubble size in highly developed turbulent flow as a function of liquid and gas flow rates and pipe diameter [102]. For a fully developed, highly turbulent flow inside a pipe, the flow can be assumed isotropic. Under this condition, the three velocity components are equal, allowing the turbulent kinetic energy of the fluid,  $E_{turb}$ , to be written as shown in Equation 1-4, where  $u_l$  is the liquid velocity, f is the friction factor of the liquid phase,  $\rho_l$ is liquid density, r, z,  $\theta$  are direction of the flow:

$$E_{turb} = \frac{1}{2}\rho_l \left(\overline{u_r}^2 + \overline{u_\theta}^2 + \overline{u_z}^2\right) = \frac{3}{2}\rho_l \overline{u_z}^2 = \frac{3}{2}\rho_l u_z^2 \frac{f}{2}$$
(1-4)

The work needed to create *a* is defined as surface free energy per unit area,  $E_{surf}$  [102].  $E_{surf}$  of bubbles with Sauter-mean diameter of  $D_{bub}$  inside a pipe with cross-sectional area of  $a_{pipe}$  can be calculated using Equation 1-3, where  $\sigma$  is surface tension. When the dispersed bubbly flow forms,  $E_{turb}$  (energy provided by the turbulent fluid) and the  $E_{surf}$  (energy needed to create bubbles) would be equal to each other as shown in Equation 1-4:

$$E_{surf} = \frac{6\sigma}{D_b} a_{pipe} u_g \tag{1-5}$$

$$\frac{3}{2}f\frac{\rho_{l}u_{l}^{2}}{2}a_{pipe}u_{l} = \frac{6\sigma}{D_{bub}}a_{pipe}u_{g}$$
(1-6)

### 1.4.2 Bubble Size Correlation Using Single-phase model

Rearranging Equation 1-4 can yield the equation for theoretical  $D_{bub}$  inside a cylindrical pipe in the following form, where *f* is friction factor:

$$D_{bub} = \frac{8\sigma u_g}{f\rho_l u_l^3} \tag{1-7}$$

For *f*, Chen et al., suggested to use the liquid pipe flow friction factor, where both  $C_f$  and  $n_f$  are arbitrary constants and *Re*:

$$f = C_f R e^{-n_f} \tag{1-8}$$

For a single-phase liquid,  $C_f = 0.046$  and  $n_f = 0.2$ . The above equation can be rewritten in form of dimensionless number in order to minimize variables, where *We* is Weber number:

$$We = \frac{8\varepsilon}{C_f R e^{-n_f}} \tag{1-9}$$

Chen et al., use of a friction factor correlation for a single liquid phase may introduce error into the equation when it is applied to two-phase flow. Garcia et al., proposed a friction factor equation for two-phase dispersed bubbly flow [107]. The mixture density, velocity, and Re, ( $\rho_{mix}$ ,  $u_{mix}$ , and  $Re_{mix}$ ) for a two-phase gas-liquid mixture are defined as follows, where  $d_{noz}$  is the ejector nozzle diameter:

$$\rho_{mix} = \rho_g \varepsilon + \rho_l (1 - \varepsilon) \tag{1-10}$$

$$u_{mix} = u_g + u_l \tag{1-11}$$

$$Re_{mix} = \frac{\rho_L u_{mix} d_{noz}}{\mu} \tag{1-12}$$
The resulting two-phase friction factor expression,  $f_{mix}$ , was correlated to be:

$$f_{mix} = 0.067 R e_{mix}^{-0.22} + \frac{16.46 R e_{mix}^{-0.99} - 0.067 R e_{mix}^{-0.22}}{[1 + (R e_{mix}/50)^{9.07}]^{0.15}}$$
(1-13)

Replacing Equation 1-6 with Equation 1-11 and plugging the friction factor expression into Equation 1-5 gives a modified Chen correlation, which will be referred as 'two-phase (2P) Chen correlation'. A comparison can be shown in Figure 1-6:



Figure 1-6. Bubble size simulation based on single (Chen) phase and two-phase friction factor

The 2P Chen correlation predicts smaller  $D_{bub}$  than the original 1P (one phase) Chen correlation. At 10%  $\varepsilon_g$  and between 20 to 30 ft/s  $u_l$  range, 2P Chen correlation predicts a 15% smaller  $D_{bub}$  compared to the 1P Chen correlation. The study to validate the differences between these two models will be addressed later in this study.

### 1.4.3 Jet Cone Angle

Upon leaving the MBE, the high-speed bubbly jet entrains the stagnant fluids from surrounding and thus increases the overall liquid flow rate inside the jet, making conventional estimation of the jet flow rate based on constant volumetric jet flow rate vastly inaccurate. Lima Neto et al (2008) investigated the stagnant fluid entrainment at high void fraction and developed for a given jet radius and axial liquid velocity [108]. However, axial liquid velocity is difficult to measure or estimate inside an industrial reactor vessel. A more recent model based on computational fluid dynamics has developed an equation to estimate the change in volumetric flow rate correlated to inlet liquid velocity, which makes it much easier to estimate the jet  $Q_l$  when entrainment is present [109].

The following simulation model assumes a Gaussian Distribution of liquid velocity inside a liquid jet.

$$u_{jet} = u_{max} e^{-50r^2/h^2}$$
 (1-14)

where  $u_{jet}$  is liquid jet velocity at any point inside a jet,  $u_{max}$  is maximum liquid velocity at the jet centerline, r is the radial position inside the jet, and h is the axial position inside jet.

Cushman-Roisin (2010) assumed jet angle from side to opposition is considered to be around 24 degrees. At any given point inside the jet, the ratio between the radii and the height of the jet would be approximately  $\tan(12^\circ)\approx 0.2$  [109, 110].

# CHAPTER TWO: MBG CAPABILITY USING *E. COLI* GROWTH AS INDICATOR

To validate the effectiveness of MBEs for microbial fermentations, a comparison experiment between the MBE and the conventional air ring sparger was conducted in the Michigan Biotechnology Institute's (MBI) fermentation pilot plant. In this comparison, *E. coli* cells inside a 300 L fermenter that was customized by MBI personnel to allow sparging by either a conventional ring sparger and an MBE. The growth rate of the cells during a sparged fermentation was measured as an indicator of the relative effectiveness of the two aeration methods.

## 2.1 Modeling Cell Growth under Mass Transfer Limitation

Batch growth of microbes exhibits multiple phases, including the lag phase, the growth phase, the stationary phase, and the death phase. During the growth phase with sufficient nutrients (carbon sources, O<sub>2</sub>, etc.) cells typically exhibit a constant doubling time, and the cell death rate is minimal. As a result, the cell number increases exponentially. During this phase, cell growth can be modeled using a mass balance on cell concentration that assumes the growth rate is proportional to the cell concentration (i.e., first order kinetics):

$$\frac{dX}{dt} = \mu_{net}X\tag{2-1}$$

where X is the cell concentration and  $\mu_{net}$  is the net specific growth rate.

### 2.2 Seed Train, Seed Source, and Broth

The cell growth inside the 300L fermenter was measured over a 24-hour period under two aeration methods: a conventional ring air sparger and MBE. The *E. coli* cell line (XL1-Blue competent cells) were grown following culturing protocols recommended for this strain [111, 112]. The original frozen cell tubes were transported using insulated container from MSU to MBI, where the cells were thawed and used to inoculate a seed train consisting of increasingly larger glass batch-culture vessels, as recommended by MBI fermentation experts. Seed-train cultures were grown in replicate to reduce the risk of the experiment being terminated by contamination or poor growth in a single vessel. Experimental details are provided in Appendix 1-1.

Figure 2-1 below is a simplified schematics of the seed train with the actual picture of the pilot-scale 300L reactor vessel that was used for this experiment.



Figure 2-1. Schematics of the seed train and the pilot-scale fermenter used for the *E. coli* cell growth experiment.

Inside the 300L bioreactor, the M9 broth was selected as the growth medium. Even though M9 growth medium does not provide higher specific growth rate compared to other medium such as LB or 2TY, the slower specific growth rate is beneficial for accurate measurement of growth rate [111]. Due to hour limits from MBI, the research team could only take samples for content measurement every hour. Thus, a 24 hours growth period with a lower growth rate was selected to ensure the growth period is long enough to fully develop the exponential phase.

#### **2.3 Fermenter and MBG Installation**

The 300L fermenter was equipped for two aeration modes. A Riverforest YJ-8 MBE (Figure 2-2 left) was installed at the bottom of the vessel for microbubble sparging. Also, an O-ring gas sparger (Figure 2-2 right) was installed inside the vessel for conventional bubble sparging. The batch cell growth kinetics resulting from the use of each aeration methods were measured in separate experiments.

The Riverforest YJ-8 MBE was connected to the main water line and the tank with NPT connector. On Figure 2-2 right, the black nozzle on the side is the gas entry point. Gas pocket is mixed with the high velocity liquid flow inside the MBE nozzle neck and shredded into microbubbles before been sparged into the main liquid body.

The O-ring gas sparger was a stainless-steel tube shaped into a circular ring at the bottom. Small holes were drilled into the bottom of the ring. Compressed air that was pumped into the top of the sparger emerged from the holes as bubble streams that rose up through the growth medium. The two spargers differed in the mechanism that controlled the bubble size. In the ring sparger, the bubble size is influenced by ratio of surface tension of the gas-liquid interface, which resists shearing of the emerging bubble off of the sparger hole, and the dynamic pressure of the liquid flowing around the bubble, which encourages shearing of the emerging bubble from the hole. The ratio of the surface tension to the dynamic pressure can be quantified using a Weber number [112].

In the MBE, intense turbulence of a gas-liquid dispersion flowing at high velocity through the MBE's throat causes disintegration of preexisting conventional bubbles into microbubbles [102].



Figure 2-2. The aeration instruments for the cell growth experiment (L: MBE, R: Ring sparger)

A schematic of the two different aeration strategies is shown in Figure 2-3. For microbubble sparging, an external centrifugal pump was used to recycle the fermentation liquid from the

reactor, through the MBE where air was added, and then into the reactor. The centrifugal pump was turned off during ring-sparger aeration, and a small stirrer (diameter < 2 cm) was installed at the bottom of the fermenter to encourage convective mixing within the reactor. In both cases, spent air was filtered and released through the top of the fermenter.



Figure 2-3. Schematic of the 300L fermenter showing the two modes of sparging

## **2.4 Measurements and Results**

## 2.4.1 Sampling Strategy

Four hours after the bioreactor was inoculated with *E. coli* culture, measurements (OD600, dissolved oxygen, glucose level, temperature) used to track the fermentation's progress commenced. Optical density of the fermentation broth at 600 nm (OD600) was measured and converted into cell density using a calibration factor: 1.0 OD600 unit value equals to  $8 \times 10^8$  cells

per ml [113]. For the first five hours, no samples were drawn since the growth is very likely to be still in the starting phase. Starting from the fifth hour, liquid content was extracted every hour for the content measurement.

Due to MBI's operating-hour constraints we were unable to take data points throughout the entire 24-hour operating period. However, sufficient data points were obtained to compare the specific cell growth rate under different sparging systems.

### 2.4.2 Growth Rate Comparisons

The following Figure 2-4 demonstrates the measured OD600 values measured for three different fermentation trials inside the 300L pilot-scale bioreactor. The OD600 values were measured and plotted as a function of time since bioreactor operation started. The previous growth equation was used to fit the curve to calculate the specific cell growth rate inside the bioreactor during different aeration modes.

Three aeration modes were investigated. The first was conventional air sparging using the ring sparger shown in the previous Figure 2-3. When using MBE to aerate the tank, a high gas flow rate (0.1 VVM or volume of air per volume of reactor per time) and a low gas flow rate (0.01 VVM) were investigated separately since the literature indicated that different  $\varepsilon_g$  has impact on  $D_{bub}$  generated by MBE [102].



Figure 2-4. Measured OD600 values for the three aeration conditions inside the fermenter

The following Table 2-4 contains calculated specific growth rate for the three different aeration mode. The final OD after an entire day of fermentation is also recorded. According to the previous Figure 2-4, only MBE at low gas flow managed to enter stationary phase while the other two aeration mode failed to achieve so. The cell growth rate for low gas flow MBE is double of the specific growth rate of air sparging.

The calculated specific growth rate for high gas flow MBE is lower than low gas flow MBE as shown in Table 2-1. At higher gas flow rate, the predicted bubble size is larger and the overall gas-liquid contact area could be smaller [102]. This could contribute to the low cell specific growth rate.

	OD after 24 hours	Growth rate (/hour)
Air sparging (0.1VVM)	0.47	0.174
Microbubble (0.1VVM)	1.7	0.24
Microbubble (0.01VVM)	2.25	0.355

Table 2-1. Calculated specific cell growth rate and OD 600 after 24 hours for the aeration methods

### 2.4.3 Glucose Level

The measured glucose level decreases as the specific cell growth propagates as it can be shown in Figure 2-5. The glucose level inside the low gas MBE was the lowest at the end of the 24-hour period. This glucose level measurement also supports the previous calculated specific growth rates. Since the low gas MBE aeration method had the highest specific growth rate, the broth should have lowest glucose content left due to consumption from cell growth. In fact, the glucose content in the broth during the low gas MBE aeration was almost exhausted by the end of the cell growth period.

The glucose level was plotted over the OD600 value instead of time to ensure a fair comparison, since the specific growth rates are different and cell concentrations are at different. The initial slopes from the starting phase look similar for all three aeration modes. Once the OD600 value surpasses 0.5, the glucose consumption rates start to differ. Since ring sparger could not deliver much gas compared to MBE, no conclusion of glucose consumption at high OD600 could be drawn from its data. The slope for the low gas MBG is much steeper than high gas MBG. This implies that at low gas condition, more glucose was consumed to produce the same number of cells.

Both the OD600 measurement and the glucose content measurement suggests that MBG at low gas flow rate managed to transfer large amount of  $O_2$  into the fermenter to sustain high cell growth. The fact that low gas MBG used more glucose to produce unit amount of cell does not change the result that low gas MBG grew the cells much faster to a much higher ceiling after 24 hours.



Figure 2-5. Glucose level within the liquid at different OD600 value

### 2.4.4 Temperature and pH

Some irregularities from the previous results could be potentially attributed to the temperature and pH irregularities during the fermentation process. These irregularities could change the cell metabolism and change the specific growth rates of the cells inside the fermenter.

The below Figure 2-6 demonstrates the pH change inside the fermenter as the cell growth

propagates. Under low  $\varepsilon_g$  MBE and ring sparger aeration, the pH of the bulk fluid inside the fermenter decreased while cell concentration increased. When using low gas MBE, the pH dropped from 7.2 to around 6 at the end of the 24-hour period. Under high gas MBE condition, however, the pH drastically reduced from 7.2 to 6.6, and then returned to 6.8.

The optimal pH for *E. coli* cell growth is between 7.5 and 6.5 [114], thus the low gas MBG aeration might produce acidic products and reduced the pH of the bulk fluid. However, this highlights the O<sub>2</sub> transfer rate provided by the low gas MBG, since this particular aeration method was capable of developing cells even outside optimal pH condition.



Figure 2-6. pH changes within the fluid for the cell growth experiment at different OD600 level

The experiment also faced temperature irregularities as shown in Figure 2-7. At the start of the experiment, the thermos jacket for the fermenter was initiated at lower temperature than the ideal growth temperature of *E. coli* cells. The ideal growth temperature for *E. coli* cells 37 °C, even though the cell is survivable between a wider range [115].

For the MBG experiments, the temperature of the thermo-jacket on the bioreactor failed to maintain the temperature to around 37 °C. This effect could also have affected the specific growth rate and introducing errors in growth rate calculations.



Figure 2-7. Temperature changes within the fluid for the cell growth experiment

# **2.5** Conclusion

The mass transfer capability of MBE was compared to conventional ring sparger inside a 300L pilot-scale bioreactor. *E. coli* cell growth was used as an indicator of  $O_2$  mass transfer rate, as higher  $O_2$  mass transfer rate usually translates into higher specific growth rate. The cell concentration, dissolved  $O_2$  content, glucose level, temperature, and pH of the media were measured to observe the cell growth.

When using high gas flow MBE, the cell grew 40 percent faster and the final cell concentration is 3.6 times higher after 24 hours than when using air sparger. When using low gas flow MBE, the cell grew at over double the speed and the final cell concentration was 4.8 times higher compared to using air sparger. The glucose content inside the media also supported the cell growth trends. Higher cell concentration corresponds to lower glucose concentration during measurements.

Some irregularities in pH and temperature might had impact the growth of the cells. Based on qualitative analysis, MBE provides better gas-to-liquid mass transfer than traditional ring sparger aeration. However, in order to implement MBE for large-scale production, further quantitative analysis is still needed.

## 2.6 Growth Media Content

The seed train protocol for the *E. coli* growth is shown here:

1. The thawed seeds were transferred to a culture plate for growth. The culture plate was left for 24 hours for growth.

2. The content on the plate is then transferred into two 5 ml test tubes. Lysogeny broth was provided by MBI and additional glucose solution was added to provide cell growth content. The test tubes were left for 24 hours for growth.

3. The content for both test tubes were measured after 24 hours. The tube with higher cell concentration was selected and the content of the tube was transferred into two 500 ml shake flasks. Additional glucose solution along with M9 broth were added to the shake flasks for cell growth. The two shake flasks were left for 24 hours for growth.

4. The content value for both flasks were measured after 24 hours. The flask with higher cell concentration was selected and the content of the flask was transferred to the 300 L fermenter. Additional glucose solution along with M9 broth were added to the fermenter as well before the final cell growth starts. The temperature for all cell growth during the seed train was 37 °C

The M9 minimal mineral medium composition can be seen in the following tables from 2-2 to 2-4. The highlighted 'M9 Salt solution' and 'trace metal solutions' compositions are recorded in the following tables as well [116]. 300 L of the aforementioned M9 minimal mineral medium was added to the fermenter, with an additional 4g/L glucose also added as the carbon source before the fermentation started.

1L M9 mineral medium		
Water	867	ml
M9 Salt solution (Table 2-2)	100	ml
20% Glucose	20	ml
1M MgSO <sub>4</sub>	1	ml
1M CaCl <sub>2</sub>	0.3	ml
biotin (1mg/ml)	1	ml
thiamin (1mg/ml)	1	ml
trace metal solution (Table 2-3)	10	ml

Table 2-2. Composition of one liter of M9 mineral medium solution

Table 2-3. Composition of one liter of M9 salt solution

1L M9 salt solution		
Water	1000	ml
Na <sub>2</sub> HPO <sub>4</sub> -2H <sub>2</sub> O	75.2	g/L
KH <sub>2</sub> PO <sub>4</sub>	30	g/L
NaCl	5	g/L
NH <sub>4</sub> Cl	5	g/L

Table 2-4. Composition of one liter of trace metal solution

1L trace metal solution		
Water	1000	ml
EDTA	5	g
FeCl <sub>3</sub>	498	mg
ZnCl <sub>2</sub>	84	mg
0.1M CuCl <sub>2</sub> -2H <sub>2</sub> O	765	μl
0.2M CoCl <sub>2</sub> -6H <sub>2</sub> O	210	μl
0.1M H <sub>3</sub> BO <sub>3</sub>	1.6	ml
1M CoCl <sub>2</sub> -6H <sub>2</sub> O	8.1	μl

# CHAPTER THREE: THEORETICAL COMPARISON OF MBG COST-EFFICIENCY BETWEEN EJECTOR (MBE) AND MODIFIED JAMESON CELL (MJC)

The previous *E. coli* cell growth study using MBE and ring sparger offered a qualitative insight on the benefit of using MBE for gas-intense operations. The next step of this study is to quantitively analyze and compare MBE with other microbubble generators.

The results presented in the literature review section suggest that microbubble-generation methods that use turbulence to form microbubble dispersions have the greatest potential of large-scale reactor applications. This section reviews the mechanisms by which turbulence results in bubble break-up to form microbubbles and describes operational properties of two types of turbulence-based MBGs (MBE and MJC) that seem most likely to cost-effectively produce microbubbles on a scale suitable for use in commercial-scale reactors. To estimate the relative cost-effectiveness of these two types, information from the literature was used to calculate estimates of the interfacial area for mass transfer generated per unit of electrical power required.

### **3.1 MBE and MJC Simulation Settings and Fundamentals**

Even though Chapter One briefly covered the mechanisms of the MBG for both MBE and MJC, there were still some more technical details that needed to be addressed to ensure a simulation with sufficient accuracy and fidelity.

The following Figure 3-1 demonstrates the simplified schematics of two reactor columns being aerated by MJC and MBE. For the MJC column, coarse bubbles rise to the top and form a foam layer. High speed liquid jet from a perforated plate would disperse the foam layer and create microbubbles. The microbubbles were then dragged downward through the column.

The MBE operation follows the setup from section 2.3. Slow liquid was boosted to high  $u_l$  and absorbs gas from the side neck of the MBE. High kinetic energy from the liquid phase shredded the gas-liquid mixture into microbubbles. The bubbly flow was then ejected into the reactor column.



Figure 3-1. Simplified schematics of MJC (L) and MBE (R) columns

### **3.2 Bubble Size Prediction and Power Consumption Prediction for MJC and MBE**

The maximum bubble diameter in a CPLJ cell is correlated to the  $Q_l$  because the bubble generation is depending on two-phase instability, which is governed by  $Q_l$  [53]. Evans et al. correlated the maximum stable bubble diameter  $(d_{max})$  inside a conventional CPLJ cell as a function of Critical Weber Number  $(We_c)$ ,  $\sigma$ ,  $\rho_l$ , and energy dissipation rate per volume ( $\phi$ ) [53]:

$$d_{max} = \left(\frac{We_c\sigma}{2}\right)^{0.6} \rho_l^{-0.2} \Phi^{-0.4}$$
(3-1)

Evans et al. also found that the  $D_{bub}$  inside a plunging column is 61% of the  $d_{max}$  [53]. A recent study Hernandez-Alvarado et al. [61] correlated the dimensionless  $D_{bub}$  inside the MJC,  $\tilde{D}_{b,m}$ , as a function of jet Weber Number ( $We_{jet}$ ), gas Froude Number ( $Fr_g$ ), dimensionless height inside the column ( $\tilde{H}$ ),  $\varepsilon$ , and dimensionless superficial velocity ( $\tilde{V}$ ) according to a recent investigation [61].

$$\widetilde{D}_{b,m} = 71.1 \widetilde{V}^{0.83} \varepsilon^{0.27} \widetilde{H}^{0.15} W e_{jet}^{-0.51} F r_g^{0.47}$$
(3-2)

MBG utilization has associated capital and operating cost with different types of MBGs. The mass transfer rate calculated from Chapter One would be a determine factor of the size and number of mass transfer vessels to achieve certain  $k_la$ . If the maximum achievable  $k_la$  is low for an MBG, then multiple mass transfer vessels would be required to achieve the target  $k_la$ . This would increase the overall capital cost of the MBG method.

For the operating costs analysis, costs such as overheads, labor costs, and raw materials costs that are not directly associated with MBG operation were assumed to be the same. The primary cost of operating the MBG investigated in this paper was the electrical power consumed by the pump to overcome the frictional pressure drop involved in generating the microbubbles. That power may be calculated using the following equation, where  $P_{pump}$  is pump power consumption,  $\Delta p$  is the pressure drop of the system, and  $Q_l$  is the liquid volumetric flow rate:

$$P_{pump} = \Delta p Q_l \tag{3-3}$$

The main pressure drop for MBE occurs in the nozzle, where the gas is added to liquid flowing at a *Re* high enough that its turbulence is sufficient to reduce bubble size to the desired diameter. That MBE friction-induced pressure drop ( $\Delta p_e$ ) can be calculated with the Garcia correlation, as it can be seen in Equation 3-4, in terms of ejector length *L*,  $d_{noz}$ ,  $\rho_l$ ,  $\rho_{mix}$ ,  $u_l$ ,  $u_{mix}$ , Fanning friction factor *f<sub>fan</sub>*, and *f<sub>mix</sub>*. [107]

$$\Delta p_e = 2f_{fan} \frac{L}{d_{noz}} \rho_l u_l^2 + 2f_{mix} \frac{L}{d_{noz}} \rho_{mix} u_{mix}^2$$
(3-4)

For the MBE, the operation mode and protocol were designed around the *E. coli* cell growth experiment. The MBE would sparge gas-liquid mixture into the reactor vessel from the bottom of the vessel. This means the MBE pump would need to work against the hydrostatic pressure. The reservoir water was stationary and the MBE pump would also need to increase the  $u_l$  to high enough value to sustain microbubble creation. Even though the MBE is short, the two-phase flow friction pressure drop was also taken into consideration. The total pressure drop across the MBE body would equal to the summation of all three components:  $\Delta P_{MBE} = \Delta p_{Hydrostatic} + \Delta p_{Friction} + \Delta p_{Kinetic}$ .

The main pressure drop inside a MJC occurs as the liquid flows through the perforated plate. The plate pressure drop  $\Delta p_m$  can be calculated using Equation 3-5 from the dimensionless pressure loss coefficient Eu,  $\rho_l$ , and  $u_l$  [117]:

$$\Delta p_m = E u \frac{\rho_l {u_l}^2}{2} \tag{3-5}$$

The value of Eu is a function of plate geometry, where both  $C_0$  and  $C_C$  are constants correlated to the perforated plate geometry, and  $\beta$  is the square root of the quotient between plate and total pore cross-sectional area,  $t_{pore}$  is the pores depth, and  $D_{pore}$  is the pore diameter [117]:

$$C_0 = 0.5 + \frac{0.178}{4(t_{pore}/D_{pore})^2 + 0.355}$$
(3-6)

$$C_c = 0.596 + 0.0031 e^{\beta/0.206} \tag{3-7}$$

$$Eu = \frac{C_0 (1 - C_C \beta^2)^2}{C_C^2 \beta^4}$$
(3-8)

For the independent variable range selection, a lower boundary 20 ft/s (6.1 m/s) superficial liquid velocity was calculated based on pipe erosion velocity inside carbon steel pipes. Pipe erosion velocity is defined as velocity where larger flow speed could cause mechanical erosion inside the pipe [118]. Pipe erosion velocity,  $u_{ero}$ , is dependent on  $\rho_{mix}$  and arbitrary pipe erosion velocity constant  $C_{ero}$  [(lb/ft-s<sup>2</sup>)<sup>0.5</sup>]for two-phase bubbly flow inside a pipe [118]:

$$u_{ero} = \frac{C_{ero}}{\sqrt{\rho_{mix}}} \tag{3-9}$$

For a continuous, non-corrosive, two-phase dispersed bubbly flow inside a pipe,  $C_{ero}$  is between 150 and 200 (lb/ft-s<sup>2</sup>)<sup>0.5</sup> [118]. At 10%  $\varepsilon_g$ , atmospheric pressure, and room temperature, the calculated  $\rho_{mix}$  is around 60 lb/ft<sup>3</sup> [118]. Based on this information, the calculated erosion velocity is around 20 to 30 ft/s for the microbubble ejector. This liquid velocity range was used as the upper and lower boundary for MBE  $u_l$ . The calculation scheme for the simulation can be seen here:



Figure 3-2. Calculation scheme for MBE and MJC cost-efficiency analysis

### 3.3 Comparison Benchmark and Algorithm

A quantitative comparison of performance properties and cost-efficiencies between MBE and MJC methods was conducted using the mathematical models described above in Figure 3-2.  $Q_l$  and  $Q_g$  were selected to be the initial independent variables. These variables were expressed in dimensionless forms as Re and  $\varepsilon_g$ . The  $u_l$  across the  $a_{pipe}$  of the ejector nozzle under a  $Q_l$  was used to calculate the Re. The  $\varepsilon_g$  was calculated by dividing  $Q_g$  by the sum of the  $Q_l$  and  $Q_g$ . The  $a_{pipe}$  for flow through the MJC's porous plate was taken to be the sum of the  $a_{pipe}$  of all pores in the plate. In simulations to compare the predicted performance properties of the MJC and MBE systems, the same  $a_{pipe}$ ,  $Q_g$  and  $Q_l$ , liquid Re, and  $\varepsilon_g$  were used for both MBGs.

The range of independent variables chosen for the simulations were chosen to match those used in published MJC studies [61]. The  $Q_l$  through the MJC column body was calculated from published data [61]. Table 3-1 contains the constant values used in the simulations.

Constant	Value	Unit
$\rho_l$	998	kg/m <sup>3</sup>
μ	9.00×10 <sup>-4</sup>	N-s/m <sup>2</sup>
σ	0.072	N/m
$\rho_g$	1.18	kg/m <sup>3</sup>

Table 3-1. Physical constants assumed for the simulations

Hernandez-Alvarado el al. used a column of square base with 0.1 m width and liquid height of 0.59 m[61]. The range of superficial column liquid velocity was 0.04 to 0.08 m/s, and that of superficial gas velocity was 0.004 to 0.02 m/s. The calculated  $Q_l$  was  $3.14 \times 10^{-4}$  to  $6.28 \times 10^{-4}$  m<sup>3</sup>/s;

the calculated  $Q_g$  was between  $3.14 \times 10^{-5}$  and  $1.57 \times 10^{-4}$  m<sup>3</sup>/s. For this simulation,  $u_l$  for the MBE was calculated based on the  $Q_l$  and  $a_{pipe}$ , and  $a_{pipe}$  along with  $d_{noz}$  were calculated by known  $Q_l$  and  $u_l$ . Liquid flow rate was converted into ejector nozzle Re, while  $Q_g$  was converted into  $\varepsilon_g$  to keep the independent variables dimensionless.

Hernandez-Alvarado et al. also reported the individual pore diameter (0.8 mm) and applicable  $We_{jet}$  value range, which was between  $5.4 \times 10^4$  and  $2.2 \times 10^5$  [61]. By the definition from Hernandez-Alvarado el al.,  $We_{jet} = u_{jet}^2 D_{pore}\rho_l/\sigma$ . The calculated  $u_{jet}$  value range based on the  $We_{jet}$  definition and pore diameter is between 6.24 and 12.6 m/s. Assuming that these  $u_{jet}$  values correspond to the same upper and lower boundaries of the published  $Q_l$  value from the same literature, the area ratio between total pore areas and the cross-sectional area of the column body was calculated based on conservation of mass flow in and out of the column. The  $\beta$  value was calculated from the pore/column area ratio. The pore depth/diameter ratio was assumed to be 1, which falls within the valid parameter range provided by Malavais., et al [117]. The calculated area ratio was that column cross-sectional area of all the pores on the perforated plate.

These parameter values were used to calculate  $D_{bub}$  values for both for the MBG systems, using Equation 1-7 for MBE and Equation 3-2 for the MJC system, as a function of the liquid phase *Re* and gas void fraction. The  $D_{bub}$  and  $\varepsilon_g$  values were then used with Equation 1-1 to calculate *a* for gas mass transfer. Pressure loss and power consumption for both the MBE and MJC systems were then calculated by solving Equation 3-4 for MBE and 3-5 for MJC. The estimated *a* was normalized by the *P* required under the same flow conditions to give a measure of costefficiency for mass transfer.

A  $k_{la}$  value of 2900 h<sup>-1</sup> that was suggested by ARPA-E as a design target for Electro-fuels fermentations [3] was used to for additional design calculations. Using Henry's Law, the ARPA-E target  $k_{la}$  is converted into an Oxygen Uptake Rate (*OUR*) to calculate the maximum theoretical reactor volume ( $V_{max}$ ) that can be sustained by the MBG:

$$V_{max} = \frac{Q_{oxygen}}{OUR} = \frac{Q_{oxygen}}{k_1 a C^*}$$
(3 - 10)

Equation 3-10 is used to calculate the theoretical maximum volume of the liquid that can be supported under certain gas and liquid flow rate conditions for both MBE and MJC methods. The results can be seen in the next section

### **3.4 Comparison Results**

Figure 3-3 is the simulated  $D_{bub}$  comparison between MJC and MBE methods. The MBE produces smaller bubbles at higher  $Q_l$ . At higher  $\varepsilon_g$ , MJC method produces smaller microbubbles. As the  $u_l$  increases, the  $D_{bub}$  produced by MBE reduces at third power due to the increase in turbulence energy provided by liquid phase. MJC-generated  $D_{bub}$  does not change much with respects to Re, but marginally increases as the  $\varepsilon_g$  increases. The strong effect of liquid Re seen for the MBE system can be attributed to Equation 1-7, which predicts that  $D_{bub}$  varies inversely with b  $u_l^3$ .



Figure 3-3.  $D_b$  correlation to liquid Re and  $\varepsilon$  for MJC and MBE method

Figure 3-4 demonstrated total predicted *a* as a function of both  $\varepsilon_g$  and *Re*. The  $D_{bub}$  results from Figure 3-3 were then used to calculate *a* value for the EJ and MJC systems as a function of *Re* and  $\varepsilon_g$  as shown in Figure 3-4. For the MBE system, *a* steadily increases as the  $Q_l$  increases but remains almost constant when altering  $Q_g$ . For MJC method, the total *a* increases as the  $Q_g$ increases. The increase in *a* is insignificant as  $u_l$  increases in MJC columns. Consistent with Figure 3-2, the MBE system is predicted to generate higher *a* value than MJC system at higher  $Q_l$  and lower gas fractions and *vice versa*.



Figure 3-4. Gas-liquid area (a) correlation to liquid Re and  $\varepsilon$  for MJC and MBE method

The primary operating cost for the two MBG systems would be electrical power required to generate the high liquid velocities required for bubble disintegration. The power requirements to overcome the  $\Delta p$  of the two systems were calculated and the result can be seen in Figure 3-5. For MBE system, a full balance on pressure-drop through the MBG using Bernoulli's principle was also conducted. Unless the *Re* was high, the power consumption for the MBE system would be higher than MJC. However, since the porous plate has a higher friction coefficient than straight pipe, the friction-induced pressure drop for MJC would increase faster than MBE system. However, it should be emphasized that these initial power calculations are approximate and further studies are required to more accurately predict the *P*<sub>pump</sub> for the two systems.



Figure 3-5. Power consumption  $(P_{pump})$  correlation to gas and liquid flow rate for MJC and MBE method

The *a* was then normalized by the  $P_{pump}$  required under the same flow conditions. Figure 3-6 demonstrates the relative cost-efficiency for mass transfer as a function of the two independent variables. The *a* per  $P_{pump}$  input as a function of  $Q_l$ . This parameter is an indicator of how 'energy-efficient' an MBG is; the total power consumption being the cost of the system while the *a* being the benefit. The higher this factor is, the more efficient a microbubble sparging system would be. MBE method has lower power consumption per unit volume comparing to MJC method at higher *Re* and lower  $\varepsilon_{g}$ . The threshold where MBE cost-efficiency surpasses is marked with a red line in Figure 3-6. Due to the significant quantity of parameters involved in this simulation, an arbitrary,  $2^{nd}$  order polynomial correlation was used to denote this threshold:  $\text{Re}_{L} = 2.84 \times 10^5 \times \varepsilon_g^2 + 1.62 \times 10^5 \times \varepsilon_g + 3.32 \times 10^4$ 



Figure 3-6. Power-efficiency  $(a/P_{pump})$  correlation to liquid *Re* and  $\varepsilon$  for MJC and MBE

Other than theoretical calculation for cost-efficiency, the scale-up difficulty of a microbubble sparging system must also be taken into consideration. An individual MBE can be modeled as an individual mass transfer cell inside a large reactor. Multiple MBEs in triangular pitch formation and in multiple layers can in theory aerate large volume of liquid inside a reactor.

To aerate industrial-scale reactor vessels, MBEs can be installed next to each other horizontally and stacked vertically inside a single, large vessel as shown in Figure 3-7. Each MBE-generated jet cone can be modeled as individual mass-transfer cell and the cells can collectively provide high  $k_{la}$  industrial-scale reactor vessel.



Figure 3-7. Vertically and horizontally stacked large scale MBE formation for fermentation.

According to Li, multiple smaller, individual MJCs with their own stir blades and perforated plates need to stack on each other to achieve a height-to-diameter ratio on par with conventional bubble columns [1]. This stacking arrangement introduces additional capital cost in column in form of additional column material and construction cost compared to the formation shown in Figure 3-7. Also, SDS was added as a surfactant to stabilize the bubble formation in both the patent (100 ppm) [1] and the bubble size estimation (10 ppm) [61] for the MJC.

Previous study on downflow column similar to MJC pointed out that liquid jet would lose penetration kinetic energy when hitting liquid phase, thus the MJC method could also face challenges when implemented on a tall column [54, 119]. Liquid jet's kinetic energy carries the microbubbles downwards, while the buoyancy force of microbubbles counter-balances the downwards force. As the kinetic energy depletes, the downwards force would also decrease, thus decreasing the downward flow speed of the microbubbles and increase the bubble residence time. As the reactive gas inside the microbubbles are expended while descending into the column, the microbubbles are left with inert gas inside. The descending microbubbles with richer inert gas could interact with the rising coarse bubbles with richer reactive gas. The microbubbles could dilute the coarse bubbles and reduce the overall mass transfer drive force of reactive gas as the bubble residence time increases. This interaction indicates that MJC MBGs can only be used on relatively short columns due to mass transfer reductions.

The bubble coalescence inside a MJC column must be taken into consideration as well. Surfactant like SDS was used to improve the bubble sizes generated inside the MJC, albeit at the cost of mass-transfer drive force [8]. Inside an MBE bubble column, the bubbles would disperse away from each other once leaving the ejector, thus reducing the chances of bubble collision and coalescence. In the patent US20140212937, the author directly indicated that the bioreactors operating with the MJC will be relatively smaller compared to conventional reactor columns [1]. This means the capital cost for implementing MJC microbubble column would be high since multiple smaller columns with their own sparging systems must be installed in order to achieve same level of  $O_2$  transfer rate of ejector types. Previous simulation results from Figure 3-6 demonstrated that the operating cost of MBE method would be lower than MJC method at higher *Re*. All factors considered, MBE method is overall the more economically attractive method to achieve higher *k*<sub>i</sub>*a* for gas-liquid reactions compared to MJC method.

# **3.5** Conclusion

Microbubble-generation methods based on intense liquid turbulence appear to offer the best combination of practicality, scalability, and cost for generating extremely high  $k_{ia}$  values in commercial-scale reactors. Mathematical models based on literature data and empirical correlations for two turbulence-based methods (MBE and MJC) allowed the relative performance characteristics of these two methods to be predicted and compared. Key dependent variables (interfacial area and power consumption required to generate that area) were calculated as function of dimensionless independent variables (*Re* and  $\varepsilon$ ).

The MBE method was predicted to generate higher interfacial area values than the MJC method at higher *Re* and lower  $\varepsilon$  values, whereas the MJC method would generate higher interfacial area values than the MBE method at lower *Re* and higher  $\varepsilon$  values. Moreover, the MBE method was predicted to achieve higher maximum  $k_ia$  value than the MJC method under the conditions studied.

For equivalent Re and  $\varepsilon$  values, the MBE method was predicted to give higher cost efficiency values (assessed as the ratio of  $k_la$  to power consumption) across the entire range of variables studied. This result suggests that turbulence generated by high Re pumping of a gas-liquid mixture through a tube is more efficient at generating microbubble dispersions than the turbulence generated by high Re pumping of a liquid through an orifice, through a gas layer, and into the surface of a continuous liquid phase.

From an operational perspective, the MBE system offers additional advantage over the MJC

system for large-scale reactors that require extremely high  $k_{la}$  values. The first disadvantage arises from the countercurrent flow of the gas and liquid feed streams in the MJC system. When the gas phase is not completely consumed in the reaction (e.g., if air is used as a source of  $O_2$ ) or if the reaction generates a gaseous product (e.g., CO<sub>2</sub>), once the reactive gas has been transported into the liquid phase, the resulting spent microbubbles containing an unreactive gas would result. Coalescence of these spent microbubbles with one another would resulting in larger, faster rising spent bubbles that would have a longer residence time than the microbubbles and thus could occupy significant reactor volume and reduce the reactor's volumetric productivity. Coalescence of spent microbubbles with the larger bubbles fed into the bottom of the MJC reactor would dilute the reactant gas in those bubbles and thereby decrease the mass-transfer driving force and volumetric mass-transfer rate. The proposed use of surfactants to reduce coalescence in MJC systems has the potential to increase production costs and complicate the product-purification process.

# CHAPTER FOUR: BUBBLE SIZE PREDICTION FOR MICROBUBBLE EJECTOR

# 4.1 Background: Experimental Validation of the Bubble Size Prediction Model and the Two-phase Friction Factor Model

The accuracy of the previous simulation results and the future implementation of MBEs during production requires the capability to predict the effective  $D_{bub}$  of microbubble dispersions generated by an MBE as a function of  $Q_l$  and  $Q_g$  values. An objective of this study was to develop that capability by measuring  $D_{bub}$  over a useful range of  $Q_l$  and  $Q_g$  values and then develop a mathematical model able to reproduce trends in the experimental results.

Optical methods based on laser diffraction [120] or image analysis of high-speed camera images [121, 122] are commonly used to measure  $D_{bub}$  in aqueous dispersions. However, given the extremely small  $D_{bub}$  and high velocity with which the microbubble dispersion exits the MBE, these approaches could not be used without specialized equipment that was not available for this study. For that reason, a novel, indirect method was developed to estimate the effective  $D_{bub}$ generated by MBE. The method entailed measuring the mass transfer rate resulting from microbubble dispersions generated by an MBE, and then calculating the effective  $D_{bub}$  that would be predicted to give the measured mass transfer rate.

### 4.2 Indirect Method to Estimate *D*<sub>bub</sub> from Dissolved Oxygen Profile

When a highly turbulent stream of microbubbles exiting an MBE is injected into lowturbulence flow environment, such as a bubble column, the  $u_l$  drops precipitously due to liquidliquid entrainment [109]. As a result, the turbulent energy intensity that disperses larger bubbles into microbubbles within the MBE is dissipated, and bubble coalescence results in a dramatic increase in  $D_{bub}$ , and corresponding decrease in a, according to Equation 1-7. For this reason, measurement of  $D_{bub}$  is difficult in low-turbulence environments unless microbubble coalescence can be prevented.

Bredwell and Worden [8] circumvented this problem adding a surfactant to the liquid phase prior to forming the microbubble dispersion. The highly polar surfactant molecules coated the microbubbles and created a local surface charge that repelled nearby microbubbles and thus inhibited coalescence. As a result, the authors were able to inject surfactant-stabilized microbubble dispersions into a flow system consisting (i.e., a bubble column) and measure the mass-transfer properties of the microbubble dispersion with minimal microbubble coalescence. They also used a mathematical model to analyze the experimental results and show that, although the surfactant layer provide the beneficial effect of stabilizing the microbubble dispersion against coalescence, it also had the detrimental effect of increasing the interphase mass transfer resistance and thereby decreasing the  $k_l$  value.[8]

In this study a novel method was developed to inhibit coalescence of a microbubble dispersion generated in an MBE and measure the mass-transfer properties of the dispersion without using a surfactant. The approach involved injecting the microbubble dispersion generated in an MBE into a flow system that consisted of a cylindrical tube having the same inner diameter as the MBE that had ports into which an oxygen probe could be mounted. As oxygen transfer occurred the dissolved oxygen concentration in the liquid phase would change, and my moving the oxygen probe to different distances down the tube, the steady-state dissolved oxygen profile (DOP) across the tube that could be measured.

Because the tube had the same inner diameter as the MBE, the degree of liquid turbulence would be expected to remain approximately the same in the MBE as across the entire flow system. As a result, the maximum stable bubble size, and hence the effective  $D_{bub}$  value, would also be expected to remain approximately constant across the flow system, even in the absence of a surfactant. Moreover, at the very high Reynolds numbers generated by an MBE, axial mixing would be expected to be negligible, allowing a relatively simple plug-flow-reactor (PFR) mixing model to be used to mathematically model two-phase flow through the system and predict the expected steady-state oxygen profile for an assumed  $D_{bub}$  value. A similar PFR model was shown to be valid for the flow system used by Bredwell and Worden. [8]

### 4.3 Mathematical Model of the DOP Inside the PFR System

The mathematical model developed to calculate the DOP across the PFR system is described below.
4.3.1 Setup and Overall Structure

The following physical phenomena were considered in developing the PFR model:

1. Both the Ideal Gas Law model and the Van der Waals model were used to simulate the effect of pressure changes on the specific gas volume of gas contained in the microbubbles.

2. Henry's Law was used to calculate the equilibrium concentration of the transferred gas at the gas-liquid interface from the bulk concentration in the gas phase.

3. Constant  $k_l$  value was assumed since the liquid and gas fluids were the same for all the experiments. For this study, air and water were selected for mass transfer operation.

4. PFR mixing model was assumed for the equipment, since the fluid velocity would be very high and flow through a narrow tube at very high Re.

5. Individual microbubbles were assumed to be spherical and identical. Since the previous  $D_{bub}$  Equation 1-5 was developed for Sauter mean bubble diameters.

6. Constant  $u_l$  inside the tube, and the relatively low  $\varepsilon_g$  does not affects  $u_l$ . This assumption is dependent on assumption 4.

Some additional physical phenomena were taken into consideration after observation on previous assumptions. These are the aspects that worth investigating to ensure model fidelity:

1. The  $D_{bub}$  might change along the way. Mass transfer between gas and liquid phase would change the bubble gas species content. Pressure change along the PFR would also change the  $D_{bub}$ depending on the equation of state.

2. The internal pressure of the bubbles changes as the bubbles travel down the pipe due to

friction induced pressure drop ( $\Delta P_{fri}$ ) and  $D_{bub}$  change.

3. The dissolved gas content,  $C_L$ , including both N<sub>2</sub> and O<sub>2</sub>, will change along the way as mass transfer occurs. This is the same for C<sup>\*</sup> of both species since the pressure change as well.

4. The O<sub>2</sub> ontent inside bubbles will change as well as the O<sub>2</sub> is being dissolved.

After examining the previous assumptions and crucial physical phenomena, a five variable ODE group was developed (Equation 4-1, 4-4, 4-9): The  $C_L$  and C<sup>\*</sup> for both O<sub>2</sub> and N<sub>2</sub>, plus internal pressure. The five initial conditions are calculated using the following protocol:

1. Initial pressure was calculated based on preset distance (Equation 4-1).

2&3. Initial  $O_2$  and  $N_2$  molar content inside an individual gas bubble, calculated using ideal gas law and later Van der Waals, initial condition 1, and calculated initial bubble radius from Equation 1-7.

4. Initial dissolved  $O_2$  was maintained to be 5 mg/L using  $O_2$  starvation method with pure  $N_2$ gas. A tank was used to collect water and  $N_2$ microbubbles were sparged in the tank to reduce the DO level in the tank till 5 mg/L.

5. Initial dissolved N<sub>2</sub> was calculated using pure N<sub>2</sub> and initial pressure with saturation.

#### 4.3.2 Flow Model Development

Two-phase pressure drop was a core component of other physical phenomena such as mass transfer drive force and bubble sizes. Equation 1-11 is the  $f_{mix}$  equation, while Equation 1-6 is the one phase  $f_{fan}$  equation. Both equations were needed to calculate the pressure drop across the MBE, but only Equation 1-11 was needed to calculate the PFR pressure drop. The usage of Equation 1-11 and Equation 1-6 was addressed in detail in section 3.2 (Equation 3-4), as MBE nozzle is only half mixture flow because the gas entry point is in the middle, while PFR is full mixture flow within the tube. The pressure drop across MBE was calculated using Equation 3-4, while the pressure drop across the entire PFR was calculated using the following equation:

$$\frac{\partial P}{\partial z} = \frac{dP}{dz} = 2\frac{\rho_{mix}v_{mix}^2 f_{mix}}{g_c d_{noz}} \tag{4-1}$$

The next equation is the individual bubble species content inside a single microbubble. The mass balance of and individual gas species *i* inside bubble with radii of  $r_{bub}$  is shown here:

$$\frac{\partial n_i}{\partial t} = k_g (4\pi r_{bub}^2) (P_i^* - P_i) \tag{4-2}$$

Since the mixing model was assumed to be PFR, the time derivative can be converted into a length derivative:

$$\frac{\partial z}{\partial t} = u_l, \frac{1}{dt} = \frac{u_l}{dz} \tag{4-3}$$

Combining the previous equations yields the following expression:

$$u_l \frac{\partial n_i}{\partial z} = k_g (4\pi r_{bub}^2) (P_i^* - P_i)$$
(4-4)

Applying ideal gas law and Henry's Law to the Equation 4-4 generated the ODE for single gas species content inside individual bubbles when using ideal gas model.  $P_{bub}$  is the internal

bubble pressure,  $V_{bub}$  is the individual bubble volume,  $n_{total}$  is the total species concentration in the bubble, *R* is gas constant, and *T* is temperature:

$$P_{bub}V_{bub} = n_{total}RT = P_{bub}\frac{4}{3}\pi r_{bub}{}^3, r_{bub} = \left(\frac{3n_{total}RT}{4\pi P_{bub}}\right)^{1/3}$$
(4-5)

However, per the previous discussion, ideal gas equation of state does not suit gas at high pressure very well. Van der Waals equation of state was then utilized in this study, where  $Van_a$  was  $1.34 \times 10^{-9}$  m<sup>3</sup>/mol and  $Van_b$  was  $3.5 \times 10^{-6}$  m<sup>3</sup>/mol:

$$V_{bub} = \frac{n_{total}R^3T^3}{P_{bub}R^2T^2 + Van_a P_{bub}^2} + n_{total}Van_b, \qquad r_{bub} = \left(\frac{3V_{bub}}{4}\right)^{1/3}$$
(4-6)

The following Henry's Law equation with Henry's constant H was used to calculate the  $C^*$  for both O<sub>2</sub> and N<sub>2</sub>:

$$C_i^* = x_i H_i P_{bub}, x_i = \frac{n_i}{\sum n_i}$$
 (4 - 7)

Combining the previous equations from 4-2 to 4-6 generated the mass balance of gas phase species inside individual bubbles. The gas phase species mass balance was conducted for both  $N_2$  and  $O_2$  inside the bubbles:

$$\frac{\partial n_i}{\partial z} = \frac{k_g (4\pi r_{bub}^2)}{u_l} (P_i - P_i^*) \tag{4-8}$$

The internal bubble pressure should be the summation of friction induced pressure drop ( $P_{fri}$ ), and Laplace pressure ( $P_{lap}$ ):

$$P_{bub} = P_{fri} + P_{lap} = 2f_{mix} \frac{\rho_{mix} v_{mix}^2 z}{g_c d_{noz}} + \frac{2\gamma}{g_c r}$$
(4-9)

Mass balance for the liquid phase was also developed between the liquid phase and the bubbles. In this case, the mass transfer equation on the right-hand side need to multiply the bubble density  $(N_{bub})$ . Also, the direction of mass transfer needs to be inversed since for this equation, the gas species are moving from gas phase to liquid phase. For this equation, no additional simplification is necessary:

$$\frac{\partial C_{L,i}}{\partial z} = \frac{4\pi k_l}{u_l} r^2 N_{bub} \left( C_{L,i} - C^* \right) \tag{4-10}$$

Likewise, there are also two variations for both  $O_2$  and  $N_2$  to fulfill the ODEs associated with initial condition 4&5.

# 4.4 Experiment Setup, Design Equations, and the Equipment Selections for the DO Experiment

# 4.4.1 Tubing

For the tubing selection, the tube roughness and  $f_{fan}$  needed to be investigated. In this particular PFR system, the  $f_{fan}$  needs to be constant at Re > 50000 to ensure consistent  $f_{fan}$  in the PFR for model accuracy. This requires a relative pipe roughness around 0.01 [123]. According to Moody chart in Figure 4-1, at d = 0.008 m, the true roughness should be larger than  $8 \times 10^{-5}$ .



The commercially available plastic tubing material's absolute roughness is around 0.002 mm or  $2 \times 10^{-6}$ . At d = 0.008 m, the relative roughness is around  $2.5 \times 10^{-4}$ .  $f_{fan}$  is around 0.02. Some common material surface roughness can be seen in the following Figure 4-2 [124].

Material	e (mm)	e (inches)	
Concrete	0.3 - 3.0	0.012 - 0.12	
Cast Iron	0.26	0.010	
Galvanized Iron	0.15	0.006	
Asphalted Cast Iron	0.12	0.0048	
Commercial or Welded Steel	0.045	0.0018	
PVC, Glass, Other Drawn Tubing	0.0015	0.00006	

Figure 4-2. Common material surface roughness in both mm and inches

The main technical requirement that needed to be satisfied when selecting the plastic tubes was the pressure and temperature ratings for this experiment. Since this experiment was conducted at room temperature, temperature rating was not a huge concern for common commercial materials. The gas delivery pressure was around 70 to 80 psi due to high  $P_{fri}$  at the start of the PFR tube with a 5 meter long and 7 mm width.

To sustain such high pressure of operation under the flow rate and tube length, a particular type of polymer tube with braid reinforcement was selected to ensure the structural integrity of the flow system was not compromised by the tubing strength under pressure. A smaller tube with similar braid reinforcement was used to connect the gas line to the regulators on the gas cylinders.

#### 4.4.2 Pumping

The pumping selection had two key parameters needed to be taken into consideration: fluid flow rate and the pressure drop. Both parameters affect the pump head and by extension the pump power requirement.

Both static and dynamic heads were calculated for the pump head. There was no static head in the PFR setup since both the pump and water tanks were on the same level. The only significant static head was the height difference between floor and water outlet. This was mitigated by elevating the tube and the pump to ensure the water entrance and exit are on the same elevation.

The dynamic head loss consisted of two parts:  $P_{fri}$  and fitting loss. Since the housing unit for sensors was a T-shape fittings along the way for probe insertion, the fitting loss needed to be

considered. The *k*-value for each straight-through T-fitting is 0.1, while it is 1.2 when it's through a side branch. The total fitting head-loss,  $h_{f}$ , was a function of velocity head and fitting loss coefficient (*k*-value)  $k_{fit}$ :

$$h_f = \frac{k_{fit} u_l^2}{2g}$$
 (4 - 11)

The friction energy head loss,  $h_{pipe}$ , due to the pipe flow was calculated using the following equations as a function of  $f_{mix}$ ,  $d_{noz}$ ,  $u_{mix}$ , tube length  $L_{tube}$ , and conversion factor  $g_c$ :

$$h_{pipe} = f_{mix} \frac{L_{tube}}{d_{noz}} \frac{u_{mix}^2}{g_c}$$
(4 - 12)

From AiChe handbook, the total power consumption (*W*) equation for a pump with head  $H_{pump}$  is a function of fluid volumetric flow rate *Q*, fluid density  $\rho$ , gravitational constant *g*, and pump efficiency  $\eta$ :

$$W = \frac{Q\rho g H_{pump}}{3.6 \times 10^6 \eta} \tag{4-13}$$

At 6.1 m/s  $u_l$ , 10%  $\varepsilon_g$ , 8 mm  $d_{noz}$ , and 6 m  $L_{tube}$ , the calculated  $H_{pump}$  was 41.44 m, the pump power was 127.7 watts, and the pump load was 0.000307 m<sup>3</sup>/s. Converting units to common pump vendor units, the units were: 135 ft, 0.17 hp, 4.86 gpm.

The calculated pump had relatively small load but very high friction head. Conventional centrifugal pump does not deliver such high head at this flow rate range. Peripheral Impeller pump fits the description better-high pump head with relatively low flow rates [125].

The selected pump was PK 100 peripheral impeller pump from *Pedrollo*. The pump motor frequency was 60 hz. The rated amperage was 6.2 A at 220 V. The pump motor was rated for 1.5

hp (1.1 kW). The rotation speed of the pump motor was 3450 rpm. To control the pump operation speed and flow rate, a variable frequency drive (VFD) was installed to control the liquid volumetric flow rate.

#### 4.4.3 Dissolved Oxygen (DO) Probe and Sensor

The DO sensor (DOS) needed for this study should to be fast reactive, small, and pressure resistant. A galvanic DOS was selected for this experiment. Galvanic DOS requires no warm-up time, and is more stable and accurate at lower DO level than polarographic probes [126]. Galvanic probe requires constant liquid flow or disturbance since  $O_2$  is actively being consumed by the galvanic cell, but that was not a problem for this study, since high  $u_l$  water flow was being flushed around the DOS constantly during the experiments. Smaller outer diameter ensures the DOS can be inserted in-line without introducing a very large side for the T-connector housing unit. A large probe side neck inside a T-connector housing unit could change the turbulence of the bubbly flow. DOS marketed for aquatic studies are required to withstand high hydrostatic pressure in natural lakes/oceans.

Atlas Lab-Grade DO Meter was selected for this experiment. This particular probe can withstand various temperature (1 ~ 60 °C), high pressure (34 atm, seven time more than max pressure for this experiment), and the maximum DO level is 100 mg/L (3 times more  $O_2$  at saturation equilibrium). The DOS was connected to a dedicated processor provided by the same vendor to compensate for pressure change throughout the experiment. The processor board was

connected to an Arduino board via co-axial cable. The connection scheme and Arduino codes were both provided by the same vendor. The galvanic cell is composed of pure silver cathode and zinc anode. The electrolyte was procured from Atlas Scientific as recommended by the vendor. The probe was then inserted into a T-connector housing unit via a dedicated ring cap to seal off the water.

#### 4.4.4 T-shaped Fitting and Barb Hose Connectors

A T-shaped fitting was chosen to connect the tubes and to insert the DOS as the housing unit. A 304 stainless steel connector with three female 1/2-inch NPT connectors was chosen as the main fitting. Two brass 5/16 inches inner diameter barb hose connector to 1/2-inch male NPT connectors were selected to connect the PFR tubes on both ends of the T-connector. Since the interior diameter of these T-connectors are larger than the flow diameters, flow through these connectors could cause some local mixing that disrupts the flow pattern inside the tube.

To avoid such disruptions, the total number of the connector for probe insertion was minimized. During the experiment, the probe would stay inside the same T-connector, while the tubes connected on both end of the connector would change to different length so the housing unit and the DOS move to a different distance from the pump via changing the PFR length at both ends of the housing unit.

The converters were attached to the T-connector, and the Atlas DOS was inserted into the connector after being secured using a dedicated ring cap to stop leakage. Additional Teflon tapes

were applied on all connectors throughout the PFR to prevent leakage under high pressure. The following Figure 4-3 shows the final product of the DO measurement housing unit using the T-connector.



Figure 4-3. The DOS connected to the main flow line with two barb hoses for the PFR

# 4.4.5 Variable Frequency Drive

A variable frequency drive (VFD) was used to control the  $Q_l$  of the pump instead of bypassing or throttling to avoid complicated fluid dynamics or fluid heating up. The VFD needs to accept the to-wall power outlet (120 V, 1 phase) and converts the current to the pump motor (220 V, 3 phases). The VFD selection was based on the parameters of the pump motor.

An ATO SKU GK3000-SP1S1-2d2 VFD was selected as the VFD between the mainline and the pump motor. 12 AWG wire that is rated for 20 amps was chosen as power line and a 20 amps circuit breaker was installed on the main-line. The pump was further grounded on the installation rack. The O-ring terminals were also welded on the connectors to ensure movement of the equipment would not risk disconnecting the lines.

## 4.4.6 Gas Flow Meter

The experiment was conducted under higher pressure compared to atmospheric pressure. The value read on the gas flow meter, which was calibrated under atmospheric pressure, does not represent the true gas flow rate. A correction equation is used here to correctly correlate the read gas flow meter values to the true gas flow rates with regards to the gauge pressure  $P_{gauge}$  and rotameter flow rate reading  $Q_{read}$  [127]:

$$Q_g = Q_{read} \sqrt{\frac{P_{pauge}}{P_{atm}}} \tag{4-14}$$

## 4.4.7 Pressure Probe

A pressure probe from the same vendor that provided the DOS was procured to measure the in-line pressure at different position inside the PFR. The measured pressure profile was used for two purposes:

1. The DOS needed pressure compensation during operation to obtain an accurate reading of the true DO value under higher pressure at a given position inside the tube. The braid that was being used to reinforce the plastic tube could have changed the surface roughness and the friction factor so it would be imprudent to use the earlier equation to estimate the pressure.

2. In order to accurately estimate the  $D_{bub}$  inside the tubes, a f was needed to calculate the turbulent energy provided. The 2P-Chen correlation used during the model development was never verified inside a narrow tube like this. An experimental validation was needed to observe whether or not Garcia correlation is suitable for this experiment.

The probe used for the pressure profile measurement used was Atlas Mega A-100 pressure probe, from the same vendor that provided the DOS. The connection was via Arduino and the code was provided by the vendor as well. The probe is inserted into the same T-connectors via an NPT adapter.

# 4.5 Mass Flow Experiment to Calibrate the VFD

Due to the employment of a VFD, the  $Q_l$  when using the pump at various VFD speed setting needed to be verified using a bucket and stopwatch method. The results are recorded in the following Table 4-1 and 4-2.

$\frac{1}{2}$ $\frac{1}$						
Time (s)	Volume (ml)	VFD Speed (hz)	$Q_l$ (ml/s)	$u_l$ (m/s)		
26.1	6980	45	267	5.97		
27.73	7440	45	268	5.99		
26.69	7100	45	266	5.94		
24.46	7310	50	299	6.68		
20.21	5990	50	296	6.62		
23.79	7010	50	295	6.58		
22.25	6615	50	297	6.64		
20.85	6800	60	326	7.28		
19.76	6480	60	328	7.32		
21.36	6860	60	321	7.17		

Table 4-1. *Q<sub>l</sub>* measurement of a 7 mm ID, 5 m long tube attached to a 7 mm MBE

Time (s)	Volume (ml)	VFD Speed (hz)	$Q_l$ (ml/s)	$u_l$ (m/s)
18.26	11390	60	624	7.94
18.2	11260	60	619	7.88
17.46	10580	60	606	7.72
16.03	9230	55	576	7.33
15.99	9200	55	575	7.33
16.49	9650	55	585	7.45
16.42	8690	50	529	6.74
15.96	8420	50	528	6.72
16.62	8670	50	522	6.64
19.88	9440	45	475	6.05
19.68	9360	45	476	6.06
19.75	9420	45	477	6.07

Table 4-2. Q<sub>l</sub> measurement of a 10 mm ID, 4 m long tube attached to a 10 mm MBE

Based on the measurement of the  $Q_l$  through the entire PFR under different VFD speed, the VFD speed for the various experiment parameters were selected. The frequencies of the pumps are selected to keep the superficial velocities of the water through the ejector nozzle to be 6, 6.6, and 7.2 m/s. The VFD speeds corresponding to these superficial  $u_l$  were recorded in the following Table 4-3.

MBE Diameter (mm)	6 m/s	6.6 m/s	7 m/s
7	45	50	60
10	45	49	54

Table 4-3.  $u_l$  measurement of a 7 mm ID, 5 m long tube attached to various MBEs

## 4.6 Mixing Model Experiment

## 4.6.1 Theory

Marshall Bredwell calculated the Peclet number (Pe) of his column and found Pe was high enough to ignore side mixing [8]. Here is the protocol to obtain Pe from measurable values according to Fogler [128].

Initially, spike the flow system (column, PFR, etc) with a concentrated salt solution to obtain the concentration curve using the concentration measurement device (i.e. conductivity probe). The time at which the signal peaks, the time interval between data collections, and the signal readings could be converted into mean residence time ( $t_m$ ) and variance ( $\sigma$ ). Using a closed-closed *Pe* correlation, variance and mean residence time can be converted into *Pe* [128].

First, collect the concentration profile C(t), which was referred as C-curve, from the photometer. Sum up the area under the C-curve to obtain the residence time distribution (RTD) curve E(t), which is now referred as E-curve. The equation for E-curve under a pulse-signal experiment is shown in Equation 4-15:

$$E(t) = \frac{C(t)}{\int_0^\infty C(t) dt}$$
(4-15)

Then, integrate the RTD curve to calculate the  $t_m$  and  $\sigma$ :

$$t_m = \int_0^\infty tE(t)dt \qquad (4-16a)$$

$$\sigma^{2} = \int_{0}^{\infty} (t - t_{m})^{2} E(t) dt \qquad (4 - 16b)$$

For a closed-closed system, the *Pe* correlation is:

$$\frac{t_m^2}{\sigma^2} = \frac{2}{Pe} - \frac{2}{Pe^2} (1 - e^{-Pe})$$
(4 - 17)

C-curve, E-curve, and the integrations were done using *trapz* function in Matlab. The final *Pe* correlation solver was calculated using *fzero* in Matlab. If *Pe* is larger than 10, it would be safe to assume that the flow has very little to no back mixing and can be categorized as PFR condition inside a column [8].

However, Bredwell experiments were conducted inside a wide column with a relatively slow  $Q_l$  across the column compared to the PFR [8]. The previous pulse-signal experiment setup was not suitable for this experiment. The  $u_l$  was so fast that the injection system could not inject the salt tracer fast enough to consider the signal to be pulse through observation. A step input was chosen and the same procedure between Equation 4-14 and Equation 4-17 was used to calculate the *Pe*. The *Pe* threshold for plug flow under a step-signal input is 78.125. The reciprocal of this number is referred as Vessel Dispersion Coefficient (VDC).

# 4.6.2 Setup

A custom-made photometer along with an appropriate dye were needed to measure the dye spike signal. The reason why photometer instead of conductivity meter was selected was because of the slow response time on the available conductivity meter. This experiment was very sensitive to the response time due to high  $u_l$  and optical signal responds faster compared to the available conductivity probe.

A generic photometer contains a light source, a sensor, and a processor to convert the sensor

signal to data that can be processed with data processing software. To ensure the measurements were accurate with little to no external interference, the light source, measurement section, and the sensor should be encased in a dark housing with on environment light shining upon it. The processor is not needed inside since it does not participate in the actual photon signal collection.

The dye selected for this experiment should meet the following criteria:

1. It should be miscible with the main liquid feed, which is water in this case to ensure no secondary phase separation deviating the measurement accuracy.

2. It should be chemically safe to dispose through main pipeline instead of a dedicated hazardous chemical waste cabinet. The total  $Q_l$  is too high and the logistics would be challenging to dispose high volume of hazardous dye water via a dedicated container.

3. It should not be reactive with  $O_2$  or  $N_2$ , since oxidation or nitration reaction could potentially change the experiment results. There will be bubbles being sparged into the pipe during mixing model experiment and the mass transfer aspects should not affect the measurement results.

After further review on existing literature, patents, and DIY instructions, the following equipment were considered for the photometer construction.

The processor kit selected was 'ELEGOO Mega 2560' from Elegoo. This vendor provided the starting kit for an Arduino processor and assorted accessories. Arduino processor could convert the measured signal into data and be processed by Matlab. The accessories include essential items such as resistors, DuPont wire, various controllers, etc.

The light sensor selected was an 818 Series Photodetector from Newport. The calibration was

done by the manufacturer before delivery. The photodetector contains a photodiode that was attached to a standard BNC connector. The BNC connector was connected to a digital Newport Power Meter (model 815). The power meter received the signal and converted the signal into analog output between 0 to 2 volts. The signal was then sent to Arduino and was recorded on the computer for analysis. An Arduino board could accept and record voltage between 0 and 5 V in 10 bits. 1024 data points are evenly distributed to represent the reading. In this case, value 0 on Arduino represents 0 V, while 410 represents 2 Vs.

The dye selected was Allura Red AC (Red 40). This type of dye was used for food coloring and was considered to be safe to dispose without using a dedicated hazardous collection vessel [129].



Figure 4-4. Allura Red AC (Red 40) chemical formula and absorption spectrum[129]

The LED light source was a 520 nm RubyLux 2nd Generation All Green LED. The diameter of the light is 4.9 cm and the power requirement is 120 V. This light was selected because it emits 520 nm light, which is close to the 504 nm optimal absorbance wavelength of Red 40.

A casing for the measurement section was produced in order to cut off environment lights and ensure the signal received by the light sensor is proportional to the light passes through the measurement medium. A circular hole with the same diameter as the LED light was cut on a piece of wood to stabilize the LED light. A channel was cut open in the middle of a wooden block to allow a transparent tube to flow through. A cuvette with bottom cut out was installed on the outside of the tube to ensure the light pathlength is straight. An additional layer was added in the middle with a small hole cut open to ensure the light pass through was both consistent and the light intensity is low enough to be recorded by the sensor. The schematic of the casing can be seen in Figure 4-5.



Figure 4-5. The schematics of the prototype photometer casing along with light source and sensor casing.

Once all the equipment were selected, the equipment for the mixing model experiment was constructed. Figure 4-6 shows the simplified schematics of the equipment used for mixing model experiment. Water was boosted inside the impeller pump for high  $u_l$  and then sent through the MBE. Since the PFR was very long, the starting pressure at MBE would be high around both the gas and dye injection point. High pressure gas inlet was connected with a pressure-resistant gas tank loaded with the dye to push the dye to the injection point after MBE. The fluid was then pushed through the PFR and detected by the prototype photometer. The optical signal for clear water and dye water were recorded using Arduino. Even though the DOS was installed during this phase, it was not used for measurement. The DOS was left in place to ensure the mixing model measured does include the DO measurement housing chamber shown in Figure 4-3 and maintains model fidelity.



Figure 4-6. Schematics for the mixing model experiment

## 4.6.3 Calibration

The analogue readings of the 815 Newport Power Meter on PC end were directly proportional to the light intensity received by the sensor. According to Beer's Law, the green light intensity passing through the Red 40 dye water should be correlated to the Red 40 concentration. By calibrating the sensor voltage/Arduino reading of the light sensor with the Red 40, a theoretical absorbance coefficient of Beer's Law could be calculated. If the sensor voltage reading follows Beer's Law closely with respects to the Red 40 dye concentration, then the sensor voltage could be used to measure the Red 40 dye concentration.

According to Beer's Law, the absorbance, A, is a function of light path distance l, dye concentration  $c_{dye}$ , and molar attunement coefficient  $\varepsilon_{att}$ .

$$A = \log \frac{I_{clear}}{I_{dye}} = \log \frac{V_{clear}}{V_{dye}} = \varepsilon_{att} c_{dye} l \tag{4-18}$$

The light received by the sensor was directly proportional to the voltage received by the Arduino board. Pure water's voltage reading was used as reference point, and the dye reading voltage was used to calculate the A. With known  $c_{dye}$  and l (the inner diameter of the tube), a  $\varepsilon_{att}$  could be calculated.

Rewrite the above equation yields exponential relationship between the sensor voltage and the dye concentration:

$$V_{dye} = V_{clear} 10^{-\varepsilon_{att} c_{dye} l} \tag{4-19}$$

Baseline light intensity and its corresponding sensor voltage  $V_{clear}$  were measured by passing through tap water into the tube. The voltage measured from the sensor was recorded as the emitted

light flux from the light source. Different  $c_{dye}$  of Red 40 were then passed through the tubes and the corresponding voltage readings are recorded and plotted against their concentration. The sensor voltage reading at different  $c_{dye}$  was recorded in Figure 4-7.



Figure 4-7. Sensor signal voltage at different Red 40 cell concentration

The calculated A of green light was plotted against the  $c_{dye}$ . The trendline is relatively straight with an R-square of 0.9971. The slope of the trend line, 8426.2 L/g, equals to the product of l (0.065m) and the  $\varepsilon_{att}$ . The calculated  $\varepsilon_{att}$  is  $12600 \pm 400$  L/mol-cm.

By plotting the signal voltage against the  $c_{dye}$ , the exponential value at log 10 of the curve was the negative product between the light path distance and the  $\varepsilon_{att}$ . The calculated A was plotted against  $c_{dye}$  in Figure 4-8 and the plot follows Beer's Law closely.



Figure 4-8. Photometer calibration of Red 40 concentration with Beer's Law

In this case, the calculated  $\varepsilon_{att}$  was 12670 L/mol-cm with R-square value of 0.9974. Both methods have good agreement with each other and has good R-square value. This indicates that 12600 L/mol-cm  $\varepsilon_{att}$  value could be used to calculate the  $c_{dye}$  inside the tube for the RTD.

#### 4.6.4 Measurement and Experiment Results

The measured *Pe* at  $u_l$  of 7.2, 6.6, and 6.0 m/s are shown in the following three figures. During the experiment, the measurements are carried out till the dye ran out. At  $u_l = 6.0$  and  $u_l = 6.6$  m/s, the concentrated Red 40 dye solution sustained 10 trials. At  $u_l = 7.2$  m/s, the dye lasted 7 experiments before ran out.

A simulated line Pe = 78.125 under step signal was constructed using the RTD equations to observe whether or not the measured RTD of the step signal mixing experiments fall under turbulent regime or not [128]. The mean and standard deviation of the measured Pe were shown in the below Table 4-4. A customized Matlab algorithm was developed to fit the curves with a calculated Pe to the RTD curve. The algorithm measures where the inflection point is on the PFR RTD using step signal. The  $t_m$  was calculated using  $u_l$  and Equation 4-15 in order to calculate the Pe.

Table 4-4. The calculated mean Pe and standard deviation for the three ul in the PFR

$u_l$ (m/s)	7.2	6.6	6
Mean	92.1	91.2	104.8
STD	11.7	12.6	12.4

At all three  $u_l$ , the measured *Pe* was higher than 78.125 (VDC <0.00128), which was the threshold for low to no back mixing according to Fogler and Levenspiele [128]. It was safe to assume the flow system for the mass transfer experiment was under plug flow conditions.



Figure 4-9a. C-curve for  $u_l = 7.3$  m/s



Fig. 4-9b. C-curve for  $u_l = 6.6$  m/s



Fig. 4-9c. C-curve for  $u_l = 6.0 \text{ m/s}$ 

As shown in the following Figure 4-10, the three measured VDC under different VFD speeds were all lower than the maximum VDC allowed for PFR model (0.0128). The curves were all



sharper than the VDC<sub>Max</sub> curve with regards to the slope around the inflection point.

Figure 4-10. Average C-curve for all three pump settings and the PFR threshold (VDC = 0.0128)

# 4.7 Pressure Drop Experiment

## 4.7.1 Theory

The mass transfer experiment requires accurate locational pressure measurement or estimation since the DOS needs to compensate for pressure during measurement. Also, the previous two-phase pressure drop model was established based on carbon-hydrate and solid flow instead of water-air [107]. This discrepancy between the literature and the PFR warrants a thorough investigation to validate the pressure drop model.

 $f_{mix}$  is crucial for this study because it was included in both  $D_{bub}$  correlation equation and  $P_{fri}$  equation. This covers both cost and benefit of MBGs. Three different  $Q_l$  and  $Q_g$  were selected.

Including pure liquid flow, the experiment has 12 different combinations of gas-liquid flow rate that are shown below:

					0	1		1	1			
Frequency (Hz)	45			50			60					
<i>u</i> <sub><i>l</i></sub> (m/s)	6			6.6				7.	2			
Qread(LPM)	0	0.35	0.7	1	0	0.35	0.7	1	0	0.35	0.7	1
<i>Qg</i> (LPM)	0	0.84	1.68	2.4	0	0.84	1.68	2.4	0	0.84	1.68	2.4
$u_g(m/s)$	0	0.33	0.67	0.96	0.00	0.33	0.67	0.96	0.00	0.33	0.67	0.96
$\mathcal{E}_{g}$	0	0.06	0.11	0.16	0.00	0.05	0.10	0.14	0.00	0.05	0.09	0.13

Table 4-5. Flow rate settings for the pressure drop experiment

The  $u_g$  and  $u_l$  were plugged in Equation 1-11 to calculate the  $Re_{mix}$  and Equation 1-12 to calculate the  $f_{mix}$  using Equation 1-13. By measuring the pressure drop of the two-phase flow inside the pipe, the slope of the pressure drops as the function of distance travelled could be used to calculate the  $f_{mix}$ . Equation 4-20 shows that the two-phase pressure drop ( $\Delta p_{mix}$ ) as a function of  $f_{mix}$ , L, pipe diameter D,  $\rho_{mix}$ , and  $u_{mix}$ :

$$\Delta p_{mix} = 2f_{mix} \frac{L}{g_c D} \rho_{mix} u_{mix}^2 \qquad (4-20)$$

The  $f_{mix}$  correlation Equation 1-13 is similar to the single-phase  $f_{fan}$  correlation Equation 1-8. If the term after the plus sign in equation 1-11 can neglected, then  $f_{mix}$  equation would be highly similar to the  $f_{fan}$  equation. A theoretical simulation of estimated  $f_{mix}$  values at the experiment range of  $Q_l$  and  $Q_g$  was conducted to assess whether or not the part after the plus sign could be ignored when calculating the  $f_{mix}$  as shown in Equation 4-21. The calculated  $Re_{mix}$  range from Table 4-5 was used as independent variables to calculate the  $f_{mix}$  using both Equation 4-21 and Equation 1-13 to observe the results.

$$f_{mix} = a_2 R e^{b_2} + \frac{a_1 R e_{mix}^{b_1} - a_2 R e_{mix}^{b_2}}{\left[1 + \left(\frac{R e_{mix}}{t}\right)^c\right]^d} \approx a_2 R e^{b_2}$$
(4 - 21)

The simulation result supported the practice of using the simplified version of Equation 4-21 instead of the complicated version of Equation 1-13 when calculating  $f_{mix}$  for the flow rates recorded in Table 4-5. The difference at any given  $Re_{mix}$  is under 1 percent compared to using the simplification method.



Figure 4-11. Theoretical simulation between the full  $f_{mix}$  form and the simplified version

#### 4.7.2 Setup

A pressure probe was modified to fit the 0.5-inch NPT port and pressure reading was recorded with a modified Arduino circuit. The pressure probe was inserted in the same 0.5-inch NPT port where the DOS inserted. An adapter that enlarges the pressure probe connector was used as connector between the probe and the port.

From observation, the data readings of the pressure gauge, regardless of digital or mechanical, were all oscillating while maintaining the same  $Q_l/Q_g$  combination. Regardless of the  $Q_l$ , probe orientation, flow rate, the pressure reading would fluctuate. The Arduino circuit could record the measured pressure data, and the average of the recorded data over 10 seconds was established as the recorded value.

The first experiment was to measure whether or not the measured pressure reading would change significantly as the liquid level inside the liquid feed tank reduces. The liquid feed tank can fit 50 gallons and the liquid level at max capacity was around 1 meter. The hydrostatic pressure was at most only 9.81 kpa, and the measured pressure drop due to friction loss was an order of magnitude larger than this value even at the slowest  $u_i$ . The pressure probe was left at 3.5 meters away from MBG. The experiment started with liquid at max level, and the liquid was being pumped into a second vessel. As the liquid level reduces, the pressure change was recorded using Arduino.



Figure 4-12. Pressure reduction as the water level drops inside tank 1.

The next phase was to measure the pressure drop inside the tube in order to calculate the f. The probe was left in same port location for the experiment, while the  $Q_l/Q_g$  combinations were changed to continuously measure the pressure readings at one location.

# 4.7.3 Experimental Result

The pressure curves at different point inside the tube were measured and the results are shown in the following figures. The  $u_l$  and  $u_g$  settings were from Table 4-5. 12 different pressure drop curves were obtained for each of the three different MBGs.



Figure 4-13. Pressure drops within PFR using the 7mm Riverforest MBE



Figure 4-14. Pressure drops within PFR using the 7mm lab-made MBE



Figure 4-15. Pressure drops within PFR using the 10 mm lab-made MBE.

From the above results shown in Figure 4-13 through 4-15, the effect of  $Q_l$  has a higher impact on the in-line pressure when the measurement point was closer to the MBE nozzle exit. When the measurement point was closer to the PFR exhaust, this effect was reduced. This was expected as the  $u_l$  has a squared effect on the  $P_{fri}$  for fluid flows inside the PFR as shown in Equation 4-20.

The slopes of the 12 curves obtained above were used to calculate the  $f_{mix}$  inside the PFR. Rearrange the Equation 4-20 yields the equation to calculate the  $f_{mix}$  using the slope (m) of the pressure drop curve:

$$\frac{\Delta P_{bub}}{\Delta z} = slope = m = 2 \frac{\rho_{mix} v_{mix}^2 f_{mix}}{d_{noz}}$$
(4 - 22)

$$f_{mix} = 2 \frac{\rho_{mix} v_{mix}^2}{md_{noz}} \tag{4-23}$$

				0			
$u_l$ (m/s)	6						
$Q_g$ (LPM)	0	0.84	1.68	2.4			
Remix	4.86E+04	5.13E+04	5.40E+04	5.63E+04			
f <sub>mix</sub>	4.63E-03	4.38E-03	4.16E-03	3.94E-03			
$u_l$ (m/s)		6	.6				
$Q_g$ (LPM)	0	0.84	1.68	2.4			
<i>Re<sub>mix</sub></i>	5.34E+04	5.61E+04	5.88E+04	6.12E+04			
$f_{mix}$	4.63E-03	4.37E-03	4.16E-03	3.99E-03			
$u_l$ (m/s)		7	.2				
$Q_g$ (LPM)	0	0.84	1.68	2.4			
Remix	5.83E+04	6.10E+04	6.37E+04	6.60E+04			
$f_{mix}$	4.42E-03	4.18E-03	3.97E-03	3.80E-03			

The calculated  $f_{mix}$  are summarized in the Table 4-6 below:

Table 4-6. Calculated  $f_{mix}$  as a function of both  $u_l$  and  $u_g$ 

After plotting the measured  $f_{mix}$  values against the estimated  $f_{mix}$  values calculated from the Equation 4-21, one discrepancy stood out: Under similar  $Re_{mix}$ , higher  $\varepsilon_g$  fluid flow produces smaller  $f_{mix}$ . According to the simulation from Equation 4-21, as the  $\varepsilon_g$  of the mixture increases, the reduction of  $f_{mix}$  should be almost linear as shown in Figure 4-16. However, according to the experimental result, the actual  $f_{mix}$  decreases significantly more than predicted as gas was introduced into the flow system.



Figure 4-16. The measured  $f_{mix}$  as a function  $Re_{mix}$  compared to Equation 4-21

Numerous previous literatures obtained mixture friction factor as a function of  $Re_{mix}$ . The form  $f = aRe^b$  is quite common among these correlations [130]. To address the significant effect of  $\varepsilon_g$  on the friction factor, an additional  $\varepsilon_g$  was appendaged in order to predict the effect of  $\varepsilon_g$  on  $f_{mix}$ . The updated  $f_{mix}$  correlation can be seen below:

$$f_{mix} = aRe_{mix}{}^{b} (1 - \varepsilon_g)^{c} \tag{4-24}$$

The  $f_{mix}$  was then processed using Matlab to fit the parameters for the three different MBEs used in this experiment. A surface fitting tool was used to accustom both independent variables. The following three surface plots depict the fitting of the experimental data on the surface plot generated using the fitted parameters a, b, and c.



Figure 4-17. Surface plot fitting of  $f_{mix}$  using Equation 4-24 for 7 mm Riverforest MBE



Figure 4-18. Surface plot fitting of  $f_{mix}$  using Equation 4-24 for 7 mm Lab-made MBE



Figure 4-19. Surface plot fitting of  $f_{mix}$  using Equation 4-24 for 10 mm Lab-made MBE

The fitting tool reported the following results and the 95% confidence interval for the parameters.

					<u>5 - 1</u>
MBG	Parameter	Average	+/-	Lower Bound	Upper Bound
River	а	0.120	0.11	0.013	0.226
	b	-0.299	0.08	-0.381	-0.217
	с	0.942	0.14	0.803	1.080
Small	а	0.127	0.17	-0.046	0.299
	b	-0.305	0.12	-0.430	-0.180
	с	0.815	0.21	0.604	1.027
Large	а	0.520	0.46	0.064	0.975
	b	-0.416	0.08	-0.494	-0.337
	с	1.224	0.21	1.010	1.437

Table 4-7. Parameter fitting results for the three MBEs using Equation 4-24.

Thus, the full modified expression of the friction factor correlation used in this study would be the following:

$$f_{mix,River} = 0.12Re_{mix}^{-0.3} (1 - \varepsilon_g)^{0.94}$$
 (4 - 25)
$$f_{mix,Small} = 0.13Re_{mix}^{-0.3} (1 - \varepsilon_g)^{0.82}$$
 (4 - 26)

$$f_{mix,Large} = 0.52Re_{mix}^{-0.42} (1 - \varepsilon_g)^{1.22}$$
 (4 - 27)

For the following experiment data fittings and simulations, Equation 4-25 through Equation 4-27 were used to calculate the internal pressure drop for the PFR when aerated using different MBEs.

For the pressure drop calculation, this  $f_{mix}$  was used for the entire length of five meters of the tube. However, when calculating the pressure drop of ejector MBGs, only half of the length was calculated using this new  $f_{mix}$  while the other half of the length pressure drop was calculated using  $f_{fan}$ . This was covered in section 3-2 with Equation 3-4. This is because the gas is injected from mid-point of the MBG, thus only second half of the nozzle is two-phase flow.

## **4.8 DOP Experiment**

With the new  $f_{mix}$  equation conducted, the mass transfer experiment could commence. DOS was inserted into the same location where pressure probe was to obtain the bubbly flow DO value. The DOS selected for this experiment required manual pressure compensation when using Arduino to processing the data.

The true performance of the manual pressure compensation needed testing in order to validate the vendor's claim on pressure compensation functionality on the DOS. The bulk fluid inside the reserve tank was flushed through the system to check whether or not the readings of the DOS at various location were proportional to their corresponding pressure. 4.8.1 Method

Reverse-osmosis (RO) water was used for this experiment to minimize the impurity in the liquid phase that could potentially change the mass transfer or bubble coalescence property. Initially, the DO level inside the bulk RO water was measured as the baseline DO level.

During the bulk fluid DO level measurement, the water pump was turned on to create some liquid flow and mixing inside the bulk fluid. The DOS used for this experiment is a galvanic probe and DO content was consumed by its galvanic cell during measurement. Liquid flow inside the tank would ensure the bulk fluid around the DOS does not exhaust DO content. The following Figure 4-20 from the DOS product brochure demonstrated that after 10 seconds without liquid movement to replenish DO content in the stagnant water, the DO level reading would decrease due to consumption and return inaccurate readings [131].



Figure 4-20. Demonstration of DOS reading reduction due to oxygen consumption During the DOP measurement, the DOS was inserted in the housing unit to record the DO

level at a certain location in the system. Since the liquid was flushing through the measurement point at very high  $u_l$ , there was no need to worry about DO consumption from the probe causing wrong readings. During operation, the commands to compensate for pressure inside the PFR was manually entered to the Arduino board to account for the drastic pressure change within the long and narrow PFR during operation.

The RO water was flushed through the water at 7.2 m/s, the highest  $u_l$  and in-line pressure this experiment would conduct under. This ensures all the possible pressure level was examined to avoid out-of-bound conditions disrupting the expectations. The RO water was flushed through the pipe for extended period of time to ensure the DO reading on the probe was stable. The pressure profile was measured in the earlier  $P_{fri}$  experiment.

During the operation, the gas valves were all turned off so the measurement was for pure liquid flow only. The liquid exhaust was moved to another tank so none of the fluid under measurement was returning to the original tank. Even though the gas valves were turned off, high speed liquid (7.2 m/s) jet hitting the liquid surface would create gas to liquid entrainment and potentially introducing fresh O<sub>2</sub> or N<sub>2</sub> to the experiment and create errors.

#### 4.8.2 DOS behavior

The measured bulk RO water concentration was 7.37 mg/L as shown in the below Figure 4-21. The pump was turned on and the DO level stabilized after being submerged in the bulk fluid at around 25 seconds. This same RO water bulk fluid was then flushed through the pipe and the



DO level was measured at various places to observe the effect of pressure on the DO level reading.

Figure 4-21. DOS stabilization when measuring RO water inside the tank

The following Figure 4-22 demonstrates the linear correlation between the distance travelled inside the tube and the probe pressure. The previous experiment on friction factor also indicated that the relationship between the distance the bubbly flow travelled and the in-line pressure at the location is linear. The DO level relationship with the distance travelled can then be converted into the relationship between DO level and pressure using Equation 4.22.



Figure 4-22. The DOS reading at various locations in the PFR using same RO water

The following Figure 4-23 demonstrates the relationship between the pressure and the DO level within the PFR. The DO values of the same fluid would increase as the liquid is being compressed at higher pressure. The relationship between DO level and the liquid pressure is almost linear with an R-square larger than 0.99.

When measuring the DO values near the MBE, the pressure would be very high due to  $P_{fri}$ . When analyzing the true mass transfer rate of the O<sub>2</sub> between gas and liquid phase, this  $P_{fri}$  would change the DO reading on the DOS when the probe was being inserted at the start of the PFR. The DOS software integrated pressure-compensation to account for this pressure change at different locations inside the flow line.



Figure 4-23. The DOS reading of the same fluid under different pressure

## 4.8.3 DOP Measurement Sequence Method

The DOP measurement was conducted under the same fluid conditions presented in Table 4-5 from section 4.7.1. The predicted pressure at a given location inside the flow system was predicted using the conclusion drawn from section 4.7. Equation 4-25 through 4-27 were used to calculate the  $f_{mix}$ , and the pressure within the tube was calculated using Equation 4-20.

The sequence of the measurements was randomly generated from statistical design in order to mitigate random errors. For this experiment, three different gas and liquid flow rates were selected. The flow rates from low to high are now described as low, medium, and high. Three replicates

were produced for the same flow rate combination to reduce the variability and to improve the confidence of the results. A total number of 27 experiment measurements (3 replicates, 3  $u_l$ , and 3  $\varepsilon_g$ ) at a particular point along the PFR was conducted before the DOS was moved to a different location.

By applying the experiment design parameters to Minitab, a random sequence order of the 27 experiments was produced. The result can be seen in the following Table 4-8. The values of the two independent variables corresponding to the flow rates were also included.

Order	$u_l (\mathrm{m/s})$	$\mathcal{E}_{g}$	Order $u_l$ (m/s)		$\mathcal{E}_{g}$	
1	Low	Medium	19	19 Medium		
2	Low	High	20 High		High	
3	Low	High	21	Medium	High	
4	Medium	Medium	22	High	High	
5	High	Medium	23	High	Low	
6	Low	Medium	24	Low	Low	
7	Low	Low	25	25 High		
8	Low	High	26 Medium		High	
9	Medium	Low	27 High		Medium	
10	Medium	High				
11	High	High	Eg			
12	High	Low		Low	5%	
13	Low	Low		Medium	10%	
14	High	Medium		High	15%	
15	Medium	Low				
16	Medium	Medium	$u_l (m/s)$			
17	Low	Medium		Low	6	
18	Medium	Low	Medium 6.6		6.6	
				High	7.2	

 Table 4-8. Sequence of measurement for PFR DOP experiment and the DOP experiment flow condition

4.8.4 Water Charging to Ensure Standard Initial DO Values.

Since the DOS was left in the same port position when measuring the DO values for 27 different DOPs, it was crucial to ensure the initial DO values of the bulk fluid were the same for all 9 port positions. Since air was used as the  $O_2$  source, it was also important to derive the bulk fluid of  $O_2$  to some extent so there could be mass-transfer drive-force between the bubbles and the water.

One method to achieve both of these goals was to sparge  $N_2$  microbubble into the bulk fluid and stop the sparging once the measured DO value of the bulk fluid was reduced to a certain value below air saturation DO value. Due to the lack of  $O_2$  inside the pure  $N_2$  microbubbles, the DO content inside the initial reservoir could be removed via liquid-to-gas mass-transfer. After considering the DOS's capability, 5 mg/L  $O_2$  content was selected as the baseline bulk fluid initial DO value. This process is now referred as 'water charging'.

The following Figure 4-24 demonstrates the schematics of the experiment during the water charging phase. A 50-gallon liquid tank filled with RO water was selected as the reservoir and was connected to the impeller pump. The impeller pump was connected to a VFD. N<sub>2</sub> gas tank and the pump outlet were connected to the MBE and the N<sub>2</sub> microbubbles were sent through the PFR. The DOS was left in place to observe the decrease in DO values of the bulk fluid. The tube was then recycled back into the same reservoir. As more and more O<sub>2</sub> molecules were removed from the bulk fluid within the reservoir by the N<sub>2</sub> microbubbles, the DO level would keep decreasing till 5 mg/L was achieved and the N<sub>2</sub> microbubbles sparging was stopped.



Figure 4-24. The schematics of the PFR during water charging operation

Once the water was charged, the DO measurement experiment could begin. The  $Q_l$  and  $Q_g$  were controlled via VFD and gas glow meter. The 27 experiments ran continuously and the DOS was moved to another location. The below Figure 4-25 demonstrates the setup for this experiment. The previous tank with charged water was used as the reservoir and the air bubbly flow was discharged into a second tank. The second tank became the new reservoir for water charging in the next experiment cycle.



Figure 4-25. The schematics of the PFR during DOP measurement

## 4.8.5 Experiment result

## 4.8.5.1 General DOP for the Three MBGs.

The DOP for the flow system when using the Riverforest MBG can be seen in the following Figure 4-26. The measured DO values increase as the bubbly flows progress down the flow line. At certain point along the line, the DO value stopped increasing due to saturation and reduction in mass-transfer drive force. As the pressure decreases, the saturated DO content in liquid phase would prevent mass transfer from occurring.

Under the same  $u_l$ , adding in more gas would increase the DO value inside the liquid phase. Similarly, under the same  $u_g$ , higher  $u_l$  would generate higher DO value. In this case, higher  $u_g$  or  $u_l$  would produce a higher  $k_l a$ .

The effect of  $u_g$  on the  $k_l a$  was also higher than the effect of  $u_l$  on it. As shown in the Figure 4-26, the three highest DOPs and  $k_l a$  values were all under high  $u_g$ . The  $k_l a$  for the highest  $u_l$  with medium  $u_g$  is still lower than the lowest  $u_l$  with high  $u_g$ .



Figure 4-26. DOP for the PFR when aerated by the Riverforest 7 mm MBE

The following Figure 4-27 shows DOP for the PFR when using the 7mm lab-made MBE. The same trend from the 7 mm Riverforest MBE DOP could be observed here. Higher  $u_g$  or  $u_l$  would increase the overall  $k_la$  of the system, while  $u_g$  has a higher impact on the  $k_la$  than  $u_l$ .



Figure 4-27. DOP for the flow system when aerated by the lab-made 7 mm MBE

The maximum measured DO value for both the lab-made and the Riverforest 7 mm MBEs are similar to each other as shown in the following Table 4-9. The highest difference for the DO value between the two different MBEs was only 7.81%.

Dissolved oxygen (M)	Riverforest	Lab-made small	Difference (%)
Low Liq-Low Gas	0.298	0.296	0.74
Low Liq-Mid Gas	0.344	0.344	0.21
Low Liq-High Gas	0.410	0.415	1.30
Mid Liq-Low Gas	0.316	0.298	5.82
Mid Liq-Mid Gas	0.356	0.366	3.06
Mid Liq-High Gas	0.422	0.455	7.81
High Liq-Low Gas	0.326	0.305	6.46
High Liq-Mid Gas	0.372	0.359	3.69
High Liq-High Gas	0.440	0.469	6.49

Table 4-9. Measured differences between the maximum observed DO value

For the larger 10 mm lab-made MBG, the general pattern of the DOP is the same as the previous smaller 7 mm MBGs' results. The DO values reached maximum at between 3 and 3.5 meters long. However, since the 10 mm tube was 1 meter shorter than the 7 mm tubes, the internal pressure inside the tube was smaller due to smaller  $P_{fri}$ . The measured DO values for the 10 mm tube were also lower than the 7 mm tubes.



Figure 4-28. DOP for the flow system when aerated by the lab-made 10 mm MBE

4.8.5.2 k<sub>l</sub>a Quantitative Analysis for the Flow Systems and Bubble Size Equation Assessment

A critical part of this study was to analyze whether Chen's correlation from Equation 1-8 and the lab-developed correlations Equation 4-24 were accurate at predicting the  $D_{bub}$  produced by the MBEs. To accomplish this, the previous model equations from section 4.3.2 were fit to the measured DOP. The specific equation utilized was the liquid phase dissolved oxygen Equation 4-10.

Matlab *lsqcurvefit* and ODE45 tools were utilized to find the optimal  $k_l$  values. The individual DOPs were fit to the predicted model and extracted from Matlab to observe whether the Equation 4-10 fits the measured DOP.

The four parameters that were being fit were the mass transfer coefficient  $k_l$ , the power factor on  $u_l$  power factor on  $u_l$ , and the initial factor for the bubble size correlations. These are the four arbitrary constants that were affecting the bubble size correlation as shown in the previous Equation 1-7. The three factors from Equation 1-7 are highlighted in red here.

$$D_{bub} = \frac{8\sigma u_g^{-1}}{f\rho_l u_l^{-3}} \tag{1-5}$$

The result of the least-square fit can be seen in the following Table 4-10 for both Chen's Equation 1-8 and the new developed 2-phase bubble size Equation 4-24.

Chen				New							
MBG	Flow	k <sub>l</sub>	U <sub>l</sub>	Ug	initial	MBG	Flow	k <sub>l</sub>	U/	Иa	initial
	LL	9.55E-04	2.81	0.94	13.98		LL	9.49E-04	2.81	0.94	14.13
	LM	1.02E-03	2.88	0.96	11.87		LM	9.96E-04	2.88	0.96	11.99
	LH	1.10E-03	2.94	0.98	9.15		LH	1.02E-03	2.96	0.99	9.25
	ML	9.94E-04	2.78	0.93	14.98		ML	9.64E-04	2.78	0.93	14.96
Riverforest	MM	9.95E-04	2.86	0.95	12.34	Riverforest	MM	9.80E-04	2.86	0.95	12.51
	MH	1.09E-03	2.93	0.98	9.72		ΜН	1.18E-03	2.89	0.96	10.32
	HL	9.71E-04	2.77	0.92	15.28		HL	2.28E-03	2.41	0.80	15.95
	HМ	9.68E-04	2.84	0.95	13.19		ΗМ	9.58E-04	2.83	0.94	13.40
	HH	1.51E-03	2.74	0.91	10.47		НН	1.01E-03	2.92	0.97	10.58
	LL	9.64E-04	2.78	0.93	14.94		LL	1.05E-03	2.75	0.92	15.42
	LΜ	1.01E-03	2.88	0.96	11.96	7mm	LM	9.93E-04	2.87	0.96	12.08
	LH	1.10E-03	2.95	0.98	9.16		LH	1.02E-03	2.96	0.99	9.22
	ML	1.01E-03	2.74	0.91	15.84		ML	9.67E-04	2.75	0.92	15.88
7mm	MM	9.93E-04	2.86	0.95	12.41		MM	9.78E-04	2.86	0.95	12.58
	MH	1.24E-03	2.87	0.96	9.29		MH	1.05E-03	2.95	0.98	9.70
	HL	9.57E-04	2.73	0.91	16.59		HL	9.43E-04	2.73	0.91	16.69
	HМ	1.00E-03	2.82	0.94	13.84		ΗМ	9.92E-04	2.81	0.94	14.05
	HH	2.92E-03	2.42	0.81	10.71		ΗН	1.02E-03	2.92	0.97	10.64
	LL	9.44E-04	2.73	0.91	16.65		LL	9.41E-04	2.73	0.91	16.68
	LΜ	9.60E-04	2.84	0.95	13.25		LΜ	9.56E-04	2.83	0.94	13.35
	LH	9.89E-04	2.94	0.98	10.05		LH	1.68E-03	2.66	0.89	10.72
	ML	9.32E-04	2.71	0.90	17.25		ML	9.28E-04	2.71	0.90	17.29
10mm	MM	9.61E-04	2.83	0.94	13.45	10mm	MM	9.55E-04	2.83	0.94	13.57
	MH	1.07E-03	2.90	0.97	10.88		MH	1.02E-03	2.91	0.97	10.91
	HL	9.33E-04	2.70	0.90	17.55		HL	9.27E-04	2.70	0.90	17.60
	HМ	9.64E-04	2.83	0.94	13.59		HМ	9.57E-04	2.82	0.94	13.71
	HH	9.99E-04	2.89	0.96	11.41		HH	1.08E-03	2.86	0.95	11.95

Table 4-10. The result of the fitted parameters for both flow models

The square-2 residual norm for all the parameter fittings were also extracted from Matlab and shown in the below Table 4-11.

Flow	2-norm residuals									
MBG	Riv	/er	Sm	nall	Large					
Model	New	Chen	New	lew Chen		Chen				
LL	0.011	0.010	0.009	0.009	0.001	0.001				
ML	0.021	0.021	0.021	0.020	0.002	0.002				
HL	0.045	0.043	0.040	0.038	0.005	0.004				
LL	0.015	0.014	0.014	0.013	0.001	0.001				
ML	0.025	0.024	0.029	0.028	0.003	0.003				
HL	0.049	0.046	0.049	0.046	0.007	0.007				
LL	0.015	0.015	0.013	0.012	0.001	0.001				
ML	0.027	0.026	0.027	0.026	0.004	0.004				
HL	0.059	0.055	0.050	0.047	0.007	0.007				

Table 4-11. The square-2 residual norm for the fitting results for both models and all MBEs

A Student T-test was conducted to observe whether the Equation 4-24 yielded significantly

different parameter fitting results when compared to the original Equation 1-8. The results can be

seen in the following Table 4-12:

 Table 4-12. Student T-test result on the fitted parameters for both old and new bubble size prediction equations

	k;	U/	Иg	initial
	0.309	0.018	0.018	0.284
DOF	52			
T-values	0.5	0.25	0.2	0.15
	0.000	0.679	0.849	1.047
	0.1	0.05	0.025	0.01
	1.298	1.675	2.007	2.400
	0.005	0.001	0.0005	0.0001
	2.674	3.255	3.488	4.002

The previous T-test results demonstrated that the mean of fitted parameters for both models were within close range of each other when modeling for the DOP using Equation 4-24. However, when the pressure inside the tube was measured, there were significant differences between using Equation 1-8 and using Equation 4-24. To maintain the consistency of the model and calculations, the  $f_{mix}$  correlation 4-24 was recommended to model.

The following figures demonstrates the DOP result of the model fitting. The dashed red lines are the simulated DOP using Equation 1-8. The solid blue lines are the simulated DOP using Equation 4-24.



Figure 4-29. The DOP fitting result for the two models using Riverforest 7 mm MBE



Figure 4-30. The DOP fitting result for the two models using lab made 7 mm MBE



Figure 4-31. The DOP fitting result for the two models using lab made 10 mm MBE

When observing the results from Table 4-10, the fitted  $k_l$  values were found to be influenced heavily by their corresponding  $\varepsilon_g$  more than their  $Re_{mix}$  as shown in Table 4-13. According to the prediction from Equation 1-7, higher  $\varepsilon_g$  would lead to larger generated  $D_{bub}$  and smaller a. However, from observations from Table 4-10, higher  $\varepsilon_g$  also contributes to higher  $k_l$  values. If lower  $\varepsilon_g$  yields higher a but lower  $k_l$ , then the overall  $k_l a$  value could increase or decrease depending on specific conditions. This unexpected 'trade-off' is detrimental to  $k_l a$  prediction.

Grouped by void								
MBG	Flow	kl	ul	ug	bubini			
	LL	9.49E-04	2.81	0.94	14.1			
Riverforest	ML	9.64E-04	2.78	0.93	15.0			
	HL	2.28E-03	2.41	0.80	16.0			
	LL	1.05E-03	2.75	0.92	15.4			
7mm	ML	9.67E-04	2.75	0.92	15.9			
	HL	9.43E-04	2.73	0.91	16.7			
	LL	9.41E-04	2.73	0.91	16.7			
10mm	ML	9.28E-04	2.71	0.90	17.3			
	HL	9.27E-04	2.70	0.90	17.6			
	LM	9.96E-04	2.88	0.96	12.0			
Riverforest	MM	9.80E-04	2.86	0.95	12.5			
	HМ	9.58E-04	2.83	0.94	13.4			
	LM	9.93E-04	2.87	0.96	12.1			
7mm	MM	9.78E-04	2.86	0.95	12.6			
	HМ	9.92E-04	2.81	0.94	14.0			
	LM	9.56E-04	2.83	0.94	13.3			
10mm	MM	9.55E-04	2.83	0.94	13.6			
	HМ	9.57E-04	2.82	0.94	13.7			
	LH	1.02E-03	2.96	0.99	9.2			
Riverforest	HL	1.18E-03	2.89	0.96	10.3			
	HH	1.01E-03	2.92	0.97	10.6			
	LH	1.02E-03	2.96	0.99	9.2			
7mm	HL	1.05E-03	2.95	0.98	9.7			
	HH	1.02E-03	2.92	0.97	10.6			
	LH	1.68E-03	2.66	0.89	10.7			
10mm	HL	1.02E-03	2.91	0.97	10.9			
	НН	1.08E-03	2.86	0.95	12.0			

Table 4-13. Parameters from Table 4-10 rearranged by void fraction independent variable

One hypothesis to explain why smaller bubbles have lower  $k_l$  is that some debris from the experiment such as oil, plastic, or metal contaminated the RO water and increased the gas-liquid membrane shell thickness [132]. To observe whether the bulk fluid used in the mass transfer experiment was contaminated, a  $k_l$  measurement was conducted to compare the bulk fluid  $k_l$  against pure RO water  $k_l$ .

## **4.9 Conclusion**

A mass-transfer experiment was conducted inside a new PFR system to observe if Fanning or mixture friction factor usage provides better prediction for  $D_{bub}$  generated with MBE. The mixing model and friction factor within the new flow system were obtained to ensure model fidelity. By measuring the DOP of the liquid within the PFR system, the  $D_{bub}$  can be indirectly calculated.

New axial-mixing and friction factor models were also developed to improve the model fidelity. Novel photometer as designed and manufactured to measure the mixing model and found that the new PFR system has little back and axial mixing. A pressure sensor was installed along the flow system to measure the pressure profile and the pressure drop along the flow system. The pressure profile was used to calculate the  $f_{mix}$  of the two-phase flow inside the PFR and to evaluate the literature  $f_{mix}$  accuracy. The literature  $f_{mix}$  model was found to be inaccurate and a new  $f_{mix}$  model was developed for this study.

The finding on the DOP was that using a new  $f_{mix}$  does not improve the model accuracy when predicting MBE  $D_{bub}$ . The lab made MBE nozzle has comparable performance when compared to the commercial MBE unit used for Chapter Two. From the parameter fitting result, smaller  $D_{bub}$ would generate higher *a* but lower  $k_l$  in the PFR when changing the fluid flow rate settings. A further study was conducted to investigate whey improving  $D_{bub}$  would actually reduce  $k_l$  and negate the benefit of smaller  $D_{bub}$  on  $k_la$ .

# CHAPTER FIVE: MASS-TRANSFER COEFFICIENT ESTIMATIONS FOR THE EXPERIMENT FLUID AND FRESH RO WATER

## 5.1 Background

The findings from the previous mass transfer experiments indicated that the measured  $k_l$  values for smaller bubbles are smaller compared to larger bubbles. A hypothesis was that some debris inside the bulk fluid was acting as barrier between gas and liquid phase [132]. To validate this hypothesis, an experiment to compare the  $k_l$  values between pure RO water and experiment bulk water was conducted.

The fundamental behind this experiment was to measure the  $k_la$  of a fluid system with same a and to calculate the  $k_la$  for the two RO water and bulk water. The fundamental equation to represent this study is the basic gas-liquid mass balance equation:

$$\frac{dC_L}{dt} = k_l a (C^* - C_L) \tag{5-1}$$

By integrating the previous equation, the theoretical DOP when aerating with constant  $C^*$  gas can be shown in the following equation:

$$\ln\left(1 - \frac{C_L}{C^*}\right) = -k_l at \tag{5-2}$$

## **5.2 Experiment Setup**

The design equation Equation 5-2. from the section 5.1 laid out multiple technical requirements for the equipment of this experiment. These technical details are being discussed in this section.

The experiment liquid should be maintained at equilibrium concentration with air before the mass-transfer occurs. Using N<sub>2</sub> microbubble or boiling to drive the initial  $C_L$  further below air equilibrium  $C_L$  could change some unknown properties of the bulk fluid. To avoid introducing further variables, air equilibrium concentration was selected as the initial  $C_L$  and no further actions were conducted on the bulk fluid once they were collected from the liquid tank before the aeration started.

Since the initial  $C_L$  was kept at air equilibrium, pure  $O_2$ , instead of air, was selected as the gas to provide mass-transfer drive force. To ensure the equilibrium concentration was truly constant, a steady  $O_2$  flow was applied to the liquid vessel's head space. The outlet of the  $O_2$  was kept at liquid level without touching the liquid. Since pure  $O_2$  is heavier than air, the incoming  $O_2$  stream would remove air inside the liquid vessel to ensure a constant equilibrium  $O_2$  concentration.

The DOS was the same DOS used in the DOP experiment in Chapter Four. Since the probe is a galvanic probe, stirring was required to prevent DO level reduction due to DO consumption via DOS. Since both the DOS and the  $O_2$  tube line need insertion on the vessel, the lid of the vessel would require openings with sufficient area.

After considering all the technical requirements, a reactor from MSU was utilized to conduct

this experiment. The schematics of the equipment and the equipment itself are shown in the following Figure 5-1.





Figure 5-1. The schematics (L) and the equipment (R) of the aeration vessel used for the  $k_{la}$  measurement experiment.

RO water and the experiment bulk water were collected and transferred to the aeration vessel. The DOS was inserted into the liquid phase and the  $O_2$  was then inserted and secured along with the DOS. The impeller was then turned on and the data acquisition started.

## **5.3 Experimental Result**

The LHS of the previous Equation 5-2 was plotted against time and the linear region in the middle of the curve was regressed. The Figure 5-2 below demonstrates the first measurement of the  $k_la$  inside the aeration vessel and the slope in the middle of the curve when using bulk fluid. The negative of the slope, which was the measured  $k_la$ , was 1.23 s<sup>-1</sup>. The similar measurements were conducted five times for both RO water and bulk fluid. The start and the end of the experimental data correspond to the beginning and the ending of the aerations. The data collected during these intervals were not corresponding to mass-transfer between pure  $O_2$  and water, since  $O_2$  was still building up in the head space. Thus, only the center part was used for linear regression calculation. Figure 5-2 shows the  $C_L$  inside the liquid phase for the first measurement with bulk fluid.



Figure 5-2. The experimental data and the linear regression at the center of the curve for the first bulk water  $k_{la}$  measurement

The below Table 5-1 shows the measured  $k_la$  values for both the RO water and the bulk fluid. A Student-T test was conducted on the two groups of data to evaluate whether these two  $k_la$  values are significantly different from each other. The average  $k_l a$  values for the RO water was  $1.47\pm0.16 \text{ s}^{-1}$ . The average  $k_l a$  values for bulk fluid was  $1.21\pm0.12 \text{ s}^{-1}$ . Both fluids had five measurements and the degrees of freedom was 8. The calculated Student-t experiment score was 2.8687, corresponding to a 98.6% confidence interval. Based on this calculation, the  $k_l a$  values for RO water and bulk fluids are different from each other. Since the measurements were conducted in the same vessel, it can be concluded that the  $k_l$  values for the RO water and bulk fluids are different from each other. This is further supported by the literature that the impurities from rust metals inside the bulk fluid might be reducing the  $k_l$  values and reducing the benefit provided by smaller bubbles. The T-connector housing unit had oil applied to the connector as corrosion prevention, and the pump was manufactured with cast iron for the pump body so these could have introduced impurities in the bulk water as the experiment progressed.

Trial	RO	Bulk		
1	1.26	1.23	t_exp	2.8687
2	1.51	1.24	DoF	8
3	1.58	1.39	CI	98.956
4	1.66	1.09		
5	1.35	1.11		
Average	1.47	1.21		
STD	0.16	0.12		

Table 5-1 Measured values and T-score calculation for the RO water and bulk fluid k<sub>l</sub>a values.

# **5.4 Conclusion**

A  $k_{la}$  measurement experiment series was conducted on the experiment water used for Chapter Four and pure RO water to observe whether or not there are significant debris within the experiment water that increased the microbubble shell thickness. The method was to measure the DOP when aerating two kinds of water within the same vessel using pure oxygen. The finding was that measured  $k_{la}$  from experiment water was lower than pure RO, thus confirming the experiment water for Chapter Four had debris and increased microbubble shell size and reduced  $k_l$  at smaller  $D_{bub}$ .

# CHAPTER SIX: LARGE-SCALE BIOREACTOR SIMULATION USING MBES

## 6.1 Background

In the previous chapters, an ejector mechanism was identified as best suited for costeffectively generating extremely high mass transfer rates in commercial-scale reactors. Then, the performance characteristics of MBE were measured and used to develop mathematical models estimate the effective microbubble size generated by MBE as a function of the key independent variables. In this chapter, those mathematical models are used to develop, for the first time, a design and scale-up algorithm for implementing MBE in commercial-scale reactors.

The gas input from one single MBE nozzle is insufficient to fully aerate an industrial-scale reactor (ISR). However, a rational scale-up strategy could be developed based on the concept of a mass-transfer cell, which is defined as the volume that would be satisfactorily aerated by a single MBE. Then arrays of MBE could be arranged into rows of mass-transfer cells, and multiple rows could be stacked as needed, as shown in the following Figure 6-1.



Figure 6-1. Sample layout of an ISR aerated using multiple layers of MBEs

Reactor system as sophisticated as such demands simulation models with substantial level of details that represents fluid dynamics inside the reactor. For this section, numerous aspects of the performance of such ISR with MBE nozzle formation were examined and compiled into intuitive results on performance that demonstrates what researchers and engineers would expect from an ISR aerated by MBE nozzle formation.

# 6.2 Jet Cone Height, Angle, and Stacking Strategy

The previous mass transfer experiment was conducted on a long, narrow PFR equipment that has the same inner diameter with the MBE. During industrial operations, an MBE would sparge bubble into a vessel with larger diameter than its inner diameter. Liquid jet entrains the stagnant fluids from surrounding and thus increases the overall liquid flow rate inside the jet [109], making estimation of the jet flow model based on constant jet  $Q_l$  vastly inaccurate.

#### 6.2.1 Stagnant Fluid Residence Time

Lima Neto et al. investigated the stagnant fluid entrainment at high void fraction and developed for a given jet radius and axial liquid velocity [108]. However, axial liquid velocity is difficult to measure or estimate inside an industrial reactor vessel. A more recent model based on computational fluid dynamics has developed an equation to estimate the change in volumetric flow rate correlated to inlet liquid velocity, which makes it much easier to estimate the jet volumetric flow rate [109].

The following simulation model assumes a Gaussian Distribution of liquid velocity inside a liquid jet.

$$u_{jet} = u_{max} e^{-50r^2/h^2} \tag{6-1}$$

where  $u_{jet}$  is liquid jet velocity at any point inside a jet,  $u_{max}$  is maximum liquid velocity at the jet centerline, r is the radial position inside the jet, and h is the axial position inside jet.

Cushman-Roisin (2010) measured jet angle from side to opposition to be around 24° [109]. At any given point inside the jet, the ratio between the radii and the height of the jet would be approximately  $\tan(12^\circ)\approx 0.2$  [109, 110].

The relationship between the  $u_{max}$  and the  $u_l$  at the MBE nozzle exit can be developed via momentum balance [109]:

$$u_{max} = \frac{5u_l d_{noz}}{h} \tag{6-2}$$

Combining Equation 6-1 and 6-2 would generate the expression for the liquid entrainment volumetric flow rate ( $Q_{ent}$ ) equation of a jet cone at a combination of  $u_l$ , h, and  $d_{noz}$ :

$$Q_{ent} = 2\pi u_{max} \int_0^\infty e^{\left(\frac{-50r^2}{h^2}\right)} r dr = 2\pi u_{max} \times \frac{h^2}{100} = \frac{u_{max}\pi h^2}{50} = \frac{u_l h \pi d_{noz}}{10}$$
(6-3)

When multiple liquid jets are propelled parallelly into the bioreactor, the regions in between the jet cones would have relatively low  $u_l$  compared to the jet zone. Through liquid entrainment, these stagnant fluids can be recycled into the jets and creates a certain 'residence time' for itself. This residence time indicates how long the organic cells inside these void zones will be out of nutrients such as O<sub>2</sub>. This residence time should be carefully designed so that the cells do not starve of nutrients.

To calculate this residence time, both the  $Q_{ent}$  and the void volume ( $V_{oid}$ ) must be calculated. For this simulation, it was assumed that the jet nozzles were packed in such way that the top of the jet cones touch each other on the edge at where 90% of the reaction gas is depleted. The jet cones were packed in a triangular pitch as shown in the below Figure 6-2.



Figure 6-2. Top-down view of a triangular pitch jet cones with tight-pack arrangement

The entrainment rate around a jet cone creates the  $Q_{ent}$  of the void region. The void region volume can be calculated via subtraction of jet cone volume from the cylinder volume of a unit cell. The volume of the cone can be determined by the following equation, where  $V_{cone}$  is the volume of the unit jet cones,  $m_g$  is the gas molar flow rate, OUR is the oxygen uptake rate and  $X_o$  is the target percentage of gas being reacted (for this simulation, the target set was 90%):

$$V_{cone} = \frac{m_g X_o}{OUR} \tag{6-4}$$

For the simulation, all the jet cones were assumed to have similar shape and size, thus the h, jet cone radius R, nozzle distance L, and subsequently the  $V_{oid}$  between the jet cones could be calculated as soon as the jet cone volume was calculated since the jet cone angle is known to be 12 degrees from center line [109]. the cylindrical and cone volume equations are shown as below for a unit cell, where  $V_{all}$  is the total volume:

$$V_{cone} = \frac{1}{3}\pi R^2 h \tag{6-5}$$

$$V_{all} = \frac{\sqrt{3}}{2}L^2h = 2\sqrt{3}R^2h \tag{6-6}$$

$$V_{oid} = 2\sqrt{3}R^2h - \frac{1}{3}\pi R^2h = \left(2\sqrt{3} - \frac{\pi}{3}\right)R^2h \tag{6-7}$$

Now that both the  $V_{oid}$  and the  $Q_{ent}$  have been calculated, the residence time of the stagnant fluid inside the void zone,  $t_{res}$ , can be calculated as:

$$t_{res} = \frac{\left(2\sqrt{3} - \frac{\pi}{3}\right)R^2h}{0.1u_{in}\pi dh} = \approx 7.7\frac{R^2}{u_{in}d}$$
(6-8)

#### 6.2.2 Pressure Drop Estimation

In order to correctly estimate the cost-efficiency of an ISR aerated by MBE (MBE-ISR), the operating cost due to pump power consumption of liquid delivery should be calculated.

When calculating the  $P_{pump}$  to operate the MBE-ISR, the same principle for calculating the MBE power cost inside the PFR also applies. Instead of  $P_{fri}$ , the hydrostatic pressure ( $P_{hyd}$ ) would be the main pressure component that was associated with the geometry of the reactor vessel. Inside the PFR, the tube was long and subsequently the  $P_{fri}$  was high. Inside an industrial-scale reactor vessel, the liquid depth would be high and the hydrostatic pressure would be high. The  $P_{pump}$  for individual MBE would be:

$$P_{pump} = \Delta P Q_l = \left(\Delta P_{hyd} + \Delta P_{fri}\right) Q_l \tag{6-9}$$

 $P_{fri}$  loss was calculated using Equation 3-4.  $P_{hyd}$  was calculated using the calculated depth of the MBE nozzle location. The detail of  $P_{hyd}$  calculation would be covered in the later section.

#### 6.2.3 Bubble Size Estimation and Interfacial Area

 $D_{bub}$  is an important indicator of the performance of a microbubble generation system since it affects both the bubble rising velocity and the *a* [34, 36]. The earlier  $D_{bub}$  prediction model results from section 4.8 indicated that using Equation 1-8 produces decent predictions on  $D_{bub}$  and  $k_{l}a$  of microbubbles exiting MBE nozzles. For the purpose of this simulation, Equation 1-8 instead of Equation 4-24 was used to predict  $D_{bub}$ .

## 6.2.4 Oxygen Transfer Rate

Gas-dissolution is usually the rate-limiting step during bioreactor operations since the rate of gas dissolving is usually lower than the rate of cell metabolism [13, 14, 133]. In this case, OUR could be approximated as mass transfer limited, where dissolved gas solubility  $C_L$  is almost zero:

$$OUR = k_l a (C^* - C_L) \approx k_l a C^* \tag{6-10}$$

The mass transfer coefficient  $k_l$  was assumed to be constant in this simulation. At mass transfer limitation, the dissolved gas concentration inside liquid phase should be approximately zero since the microorganisms are consuming nutrients faster than the gas inflow replenishes it.

The equilibrium gas concentration  $C^*$  is dependent on the inner pressure of the gas bubbles, which is further dependent on the position of the bubble inside the column due to the influence of depth on hydrostatic pressure. According to Henry's law,  $C^*$  for O<sub>2</sub> is proportional to both the total pressure  $P_{total}$  and the O<sub>2</sub> content ( $y_{O2}$ ) inside the gas.

$$C^* = y_{o_2} H_{en} P_{total} \tag{6-11}$$

The liquid entrainment would increase the total  $Q_l$  as the jet progresses inside the cone. Meanwhile gas consumption and pressure change would alter the  $C^*$  as the jet progresses since reactive gas is being consumed. These two factors would change the *a* as the jet rises as well.

The total pressure at the exit of the nozzle and the termination point of the jet cone would be different due to the difference in  $P_{hyd}$  and depletion of reactive gas. This would change  $C^*$  as the jet progresses. Since both *a* and  $C^*$  are not constant across the jet cone, Equation 6-10 was no longer an accurate representation of the actual *OUR* inside the jet cone. This issue was resolved

by utilizing an analog to the log mean temperature difference to represent the mass transfer drive force difference along the jet cone:

$$\overline{aC^*} = \frac{a_{top}C^*_{top} - a_{bottom}C^*_{bottom}}{\ln\left(\frac{a_{top}C^*_{top}}{a_{bottom}C^*_{bottom}}\right)}$$
(6 - 12)  
$$OUR_{actual} = k_l \overline{aC^*}$$
(6 - 13)

The following Figure 6-3 provides an illustration of the log mean mass transfer drive force for a triangular pitch formation.



Figure 6-3. Sample triangular pitch ejector cones and the log mean mass transfer boundary

The  $y_{O2}$  at the jet bottom (exit of the nozzle) was the O<sub>2</sub> content of the inlet gas, which was 0.2 for air for the purpose of this simulation. At the top of the jet cone,  $y_{O2}$  would decrease due to dissolution of gas inside the liquid phase. At 90% exhaustion of reactive gas, the  $y_{O2}$  value at the top of the jet cone would be:

$$y_{o_2,top} = \frac{y_{o_2} - 0.9y_{o_2}}{1 - 0.9y_{o_2}} = \frac{0.1y_{o_2}}{1 - 0.9y_{o_2}}$$
(6 - 14)

The  $\varepsilon_g$  at the top of the jet cone would decrease since  $Q_l$  would increase due to entrainment and  $Q_g$  would decrease due to gas consumption. Applying Equation 6-3 and the denominator of Equation 6-14 into void fraction equation would yield the  $\varepsilon_g$  at the top of the jet cone:

$$\varepsilon_{top} = \frac{Q_g (1 - 0.9 y_{o_2})}{Q_l + 0.1 u_{in} \pi dh} \tag{6-15}$$

The final expression for the top and bottom  $aC^*$  in Equation 6-12 would look like the following, where:

$$a_{bottom}C^*_{bottom} = \frac{6u_l}{d_{bubble}u_g}y_{o_2}H_{en}P_{bottom}$$
(6 - 16)

$$a_{top}C^*_{top} = \frac{6}{d_{bubble}} \frac{Q_g(1-0.9y_{o_2})}{Q_l+0.1u_{in}\pi dh} \frac{0.1y_{o_2}}{1-0.9y_{o_2}} H_{en}P_{top}$$
(6-17)

## 6.2.5 Dissipation Energy

Ejector style microbubble generator relies on liquid phase turbulence energy to create microbubbles [102, 134]. The same turbulence could cause cell damage and reduce the overall production inside the bioreactors [135, 136]. This means any optimization processes should take the level of cell damage into consideration since increasing turbulence without taking cell damage into consideration would be counter-productive.

Different research groups have studied the source and effect of shear damage on various kinds of cells inside different types of devices. Millward & Bellhouse (1994) investigated turbulenceinduced damage on Mammalian cell under laminar flow regime inside a membrane bioreactor [137]. Li et al. (2011) studied the effect of temperature on the cell viability reduction due to shear damage [138]. It was discovered that increasing medium temperature would make the cells more susceptible to shear damage. The effect of stirring-induced shear on cell viability has also been investigated before on insect cells [136]. Multiple papers were also published on how to evaluate shear stress inside various reactor vessels. Sánchez Pérez el al. (2006) developed a theoretical correlation of stirrer shear rate inside a STR for both laminar and turbulent flow regimes [135]. Liu et al. (2013) investigated the shear stress distribution, turbulent kinetic energy, and energy dissipation rate inside a two-phase dispersed bubbly flow inside confined nozzle spaces and developed, and discovered that the turbulent energy is the main cause to the cell death inside a bioreactor [139]. However, Liu et al. (2013) did not develop an explicit equation that's suitable to estimate the level of turbulence the microorganisms are undergoing inside ejector nozzles.

Reichmann et al. utilized a way to estimate the dissipation energy exerted on the fluids when investigating two-phase reactions inside bioreactors [140]. The same equation was used in other studies on bioreactors that involves in high turbulence flow conditions [141, 142]. The dissipation energy  $E_{diss}$  is a function of  $\Delta p$ ,  $Q_l$ ,  $\rho_l$ , and ejector volume  $V_{ejector}$ .

$$E_{diss} = \frac{\Delta P Q_L}{\rho_L V_{ejector}} \tag{6-18}$$
#### 6.2.6 Simulation Setting

For the following simulation, the *OUR*,  $P_{pump}$ , and  $E_{diss}$  were evaluate for a bioreactor via simulation using Matlab as calculation and plotting software. Combinations of  $u_l$  and  $d_{noz}$  were selected as independent variables. The range of  $u_l$  was between 20 ~ 30 ft/s and the range of  $d_{noz}$  selected was between 1 ~ 10 cm. In order to maintain the jet nozzle formation shown in Figure 6-2, the jet cone size, and subsequently jet height and distance between jet nozzles, were kept as a constant inside the bioreactor.

At the bottom of the bioreactor, the  $\varepsilon_g$  was set to be 0.1. The  $V_{cone}$  and h for the mass transfer unit cells was calculated using Equation 6-4 with known  $m_g$  and OUR.  $m_g$  was a known variable due to constant  $\varepsilon_g$  at the bottom. Matlab *fzero* function was used to calculate OUR and jet height via the following method:

1. Assume the jet cone *OUR* is the *OUR* calculated using Equation 6-10 based on the flow conditions at the exit of the ejector nozzles. Use this to calculated an initial guess jet height required by *fzero* function.

2. Assume a new jet height value  $h_{jet}$ . The total pressure used in Equation 6-11 for the bottom and top of the jet cone are with this assumed jet height in a vessel with height  $H_{tank}$  would be:

$$P_{bottom} = \rho g H_{tank} \tag{6-19}$$

$$P_{top} = \rho g \big( H_{tank} - h_{jet} \big) \tag{6-20}$$

3. Plug in Equation 6-19 and 6-20 into Equation 6-16 and Equation 6-17 will yield the components for Equation 6-12 and 6-13. In this way the *OUR* could be calculated based on

Equation 6-13

4. With both *OUR* and  $Q_g$  calculated, the  $V_{cone}$ , and subsequently  $h_{jet}$  could be calculated. The calculated jet cone height needed to match the assumed  $h_{jet}$  from step 2. In Excel this can be accomplished via Goalseek function. In Matlab this was accomplished with *fzero* function.

5. Consolidate step 2 ~ 4 to formulate the equations into a one-variable equation where  $h_{jet}$  was the only unknown. Use the initial guess calculated from step 1 for *fzero* function to calculate the actual  $h_{jet}$ . The calculated  $h_{jet}$ , based on the chosen pair of  $u_l$  and  $d_{noz}$ , was the true  $h_{jet}$  of all the mass transfer units inside the bioreactor for the chosen independent variables.

6. As the nozzle formation progresses upwards inside a large-scale bioreactor, the  $u_g$  would change as well due to  $P_{hyd}$  change as the liquid gets shallower. For the upper-level jet nozzles, solve for  $u_g$  with a different pressure to compensate for this change.

7. With  $u_g$  and  $h_{jet}$  calculated, an accurate estimation of OUR, W, and  $E_{diss}$  could be obtained. Preliminary results obtained via this method indicated that  $D_{bub}$  does not change much as at different pressure, which supports the previous assumption.

8. The number of jet nozzles per level was calculated by rounding down the ratio between total bioreactor cross-sectional area and the cross-sectional area of maximum jet cone. This ensures the entire column as covered with mass transfer units in order to maximize the production rate of the bioreactor.

9. *OUR* and power consumption values were summed for the entire bioreactor vessel. The simulation software was capable of calculating cumulative dissipation energy, albeit instantaneous

maximum dissipation energy inside the bioreactor was more representative to the maximum strain cells undergoes. The reason why maximum was chosen was because the summation of the  $E_{diss}$  does not reflect on the level of strains microorganisms undergo individually.

The following values for chosen for the constants that were added for this section

Variable	Value	unit
Hen,298: Henry's constant at 298 Kelvin	1.28E-05[143]	mol/m <sup>3</sup> -Pa
$k_l$ : mass transfer coefficient	0.0001	m/s
l/d: nozzle length to diameter ratio	5	
$P_{atm}$ : atmospheric pressure	101325	Pa
<i>R</i> : gas constant	8.314	m <sup>3</sup> -Pa/K-mol
<i>T</i> : Temperature	300	K
<i>V<sub>reactor</sub></i> : reactor volume	500	m <sup>3</sup>
H/D: height/diameter ratio of the bioreactor	2	
$X_{O2}$ : oxygen reaction extent	0.9	
$y_{O2}$ :oxygen content of inlet gas	0.2	
$\sigma$ : liquid surface tension	0.072[144]	N/m

Table 6-1. The new variable sheet for large-scale simulation

#### **6.3 Simulation Results**

The following Figure 6-4 shows the  $D_{bub}$  corresponding to different  $u_l$  and  $d_{noz}$ . For the chosen range of independent variables, the bubbles produced were all on micrometer scale.  $D_{bub}$  sizes increase as the  $u_l$  decreases, mostly due to the reduction in available turbulence energy.  $d_{noz}$  does not change the bubble size much.



Figure 6-4. Theoretical  $D_b$  generated inside the MBE-ISR

Figure 6-5 demonstrates that the calculated  $h_{jet}$  as a function of both  $u_l$  and  $d_{noz}$ . Increasing  $u_l$  marginally decreases the calculated  $h_{jet}$ . Increasing  $d_{noz}$  would raise the calculated  $h_{jet}$  significantly since the  $\varepsilon_g$  at the bottom of the bioreactor was a constant,  $u_g$  and  $m_g$  would increase significantly, extending the size of the cone the two-phase flow could sustain.



Figure 6-5. Theoretical  $h_{jet}$  as a function of both  $u_l$  and  $d_{noz}$  inside the MBE-ISR

The  $P_{pump}$  at various level of  $u_l$  and  $d_{noz}$  is shown in Figure 6-6 The total W of the entire bioreactor suddenly increases to a higher level as  $d_{noz}$  decreases. This sudden increase is due to the addition of a new layer of nozzles inside the bioreactor. The W of the system decreases marginally as the  $d_{noz}$  decreases until the sudden bump due to increase in number of nozzles. As the  $u_l$  increases, the total W increases as well due to higher  $Q_l$ .



Figure 6-6. Total power consumption as a function of both  $u_l$  and  $d_{noz}$  inside the simulated MBE-ISR

Similar trends in Figure 6-7 can be observed for the jet nozzle number as a function of  $u_l$  and  $d_{noz}$ . Due to the increase in  $d_{noz}$ , the total  $Q_g$ , along with the  $V_{cone}$ , would increase. This would reduce the number of nozzles needed to aerate the entire vessel, as shown in Figure 6-7 Raising  $u_l$  also marginally increases the total number of nozzles as well. This is surprising mainly because as  $u_l$  surges,  $u_g$  and  $Q_l$  should increase as well at constant bottom  $\varepsilon_g$ . This trend should reduce the total number of jet nozzles as the  $V_{cone}$  should increase, but the simulation result in Figure 6-7 demonstrates otherwise. A conclusion that could be drawn is that increasing  $u_l$  at constant  $d_{noz}$  inside a tight pack MBE-ISR would reduce the  $V_{cone}$  due to entrainment-induced mass drive force change.



Figure 6-7. Total number of MBE nozzles needed to aerate the entire MBE-ISR as a function of both  $u_l$  and  $d_{noz}$ 

Figure 6-8 is the calculated  $t_{res}$  as a function of  $u_l$  and  $d_{noz}$ . This parameter indicates how long the microorganisms would be deprived of nutrients until they are recycled into the jet cones. Compare this result with the total number of nozzles in Figure 6-7, it was found out that adding a new layer of nozzles as the  $V_{cone}$  decreases would increase the  $t_{res}$ . Increasing  $u_l$  would marginally decrease the  $t_{res}$ . Two lines were marked out on Figure 6-8 to indicate where the sudden drops in  $t_{res}$  occur. These two lines describe the  $u_l$  and  $d_{noz}$  combination that produces the exact mass-transfer unit cell that fills up the entirety of the BIG reactor with no wasted head-space. This is the optimal  $u_l$  and  $d_{noz}$  combination the BIG reactor should be operating at, since this combination would minimize the wasted space.

The first step would be selecting the layer of nozzles wanted for the reactor. The first red line where  $u_l = 215.15 d_{noz}$ -11.988 corresponds to two layers. The other  $u_l = 315.29 d_{noz}$ -11.736

corresponds to three layers. The second step would be selecting a  $d_{noz}$ . This design decision is usually influenced by the available MBE nozzles on the market.

The last step would be calculating the  $u_l$  based on these two equations. This way, at 10%  $\varepsilon_g$ , the selected  $u_l$  and  $d_{noz}$  combination would create mass transfer unit cell jet cones that fill up the entirety of the MBE-ISR with no wasted head space.



Figure 6-8. Stagnant fluid residence time inside the MBE-ISR as a function of both  $u_l$  and  $d_{noz}$ 

Figure 6-9 is the maximum  $E_{diss}$  inside the bioreactor at any given  $u_l$  and  $d_{noz}$ . For the present work, only the maximum level of  $E_{diss}$  was calculated. Smaller  $d_{noz}$  has higher  $E_{diss}$  due to higher pressure drop at inside the bioreactor. Increasing the  $u_l$  only marginally increases  $E_{diss}$ .



Figure 6-9. Maximum  $E_{diss}$  as a function of both  $u_l$  and  $d_{noz}$  inside the MBE-ISR

Figure 6-10 is the calculated total OUR as a function of  $u_l$  and  $d_{noz}$ . The total OUR increases at lower  $d_{noz}$ . OUR also increases significantly as  $u_l$  increases. This tendency in OUR favors scale up  $Q_l$  during continuous operation since higher  $Q_l$  provides higher OUR for a MBE-ISR. Both OUR and  $P_{pump}$  shares the same trend but on a different scale so it was difficult to predict whether or not a combination of independent variables would be cost efficient. To visualize this effect, a new parameter,  $OUR/P_{pump}$ , was introduced as an indicator to the cost efficiency of the system, where OUR is the benefit, and W is considered to be the cost.



Figure 6-10. Total OUR inside the bioreactor as a function of both  $u_l$  and  $d_{noz}$  inside the ISR

Figure 6-11 demonstrates the cost-efficiency for a BIG bioreactor by plotting  $OUR/P_{pump}$  as a function of both  $u_l$  and  $d_{noz}$ . As a new layer of nozzles was added to the system due to shrinking  $V_{cone}$ , the cost efficiency value undergoes a marginally small drop. This trend indicates that adding a newer layer of nozzles reduces the cost-efficiency somehow, mainly due to a newer layer just aerating a small volume at the top of the column was not very beneficial.



Figure 6-11. Total  $OUR/P_{pump}$  inside the bioreactor as a function of both  $u_l$  and  $d_{noz}$ 

Power per reactor volume is also an important factor that needs to be concerned with when designing a large-scale bioreactor. Figure 6-12, shows the estimated values of power per volume under different conditions. This value shares the same trend with power consumption value.



Figure 6-12. Power density inside the bioreactor as a function of both  $u_l$  and  $d_{noz}$ 

## CONCLUSION

This study started to explore whether or not microbubble ejector is a cost-effective method to improve the gas to liquid mass-transfer rate. *E. coli* cell growth experiment inside a 300L pilot scale reactor vessel confirmed MBE's aeration rate advantage over conventional ring sparger and stirrer combination strategy. MBE managed to grow *E. coli* cells at double the growth rate compared to the conventional stirrer.

Afterwards, a critical literature study was conducted to study the mechanism of various MBGs and explored the cost-efficiency of a modified Jameson Cell system from Lonza Tech and the microbubble ejector used for this study. The theoretical study showed that MJC system has better cost-efficiency at higher gas void fraction, while the ejector has higher cost-efficiency at higher *Re*. The intersection threshold of which one particular MBG is better than the other one was also developed and can be helpful to engineers during decision making phase when designing gas-intense operations.

Following up the theoretical simulation of microbubble ejector performance was the experiments to validate the actual performance capability of a microbubble ejector. The most aspect of the MBE performance that requires validation was the produced microbubbles'  $D_{bub}$  upon leaving the ejector nozzle. A new two-phase friction model was developed and this following study was to investigate whether the two-phase friction model represents the true bubble size better than the old one-phase Fanning friction model. Due to the technical challenge to measure the bubbles

directly using optical signal, an alternative indirective bubble size measurement method was developed. By sparging air bubbles into oxygen-deprived water, the DOP can be used to estimate the mass-transfer characteristics of the microbubbles. By working backwards from the mass-transfer properties of the bubbly flow, the  $D_{bub}$  of the microbubbles produced by MBE can be indirectly measured. This indirectly measured microbubble diameter could be used to verify the aforementioned bubble size correlation equation.

To ensure the DOP actually represents the mass transfer properties of fresh microbubbles leaving the MBE, the flow system was modified. The key to ensure the full DOP behaves as if the bubbles just left MBE, the flow system was set to be a long tube so the superficial liquid velocity and the kinetic energy associated with it could be constant. As long as the liquid phase kinetic energy is constant, the bubble sizes should be maintained to be constant.

Since the equipment was different from existing models, the actual mixing model and friction factor needed to be actually measured instead of deducted using existing literature. A custom photometer was designed and manufactured to measure the Residence Time Distribution of the flow system to calculate the mixing model of the custom system. A photometer was used because normal conductivity probe and salt tracer used for RTD measurements are too slow to react to the high-speed fluid in the custom flow system. The conclusion was that PFR flow model fits the new flow system's RTD.

Pressure drop profile was used to calculate the measured two-phase friction factor; it was discovered that the Garcia correlation did not fit this experiment's data well. A new pressure drop

correlation was developed to account for the effect of void fraction on two-phase friction factor. The new correlation was used in the following mass-transfer experiment to account for the change in mass-transfer drive force in the mass balance between bubbles and the liquid phase.

A collection of mass-transfer equations was developed to estimate the change in the DO value in the liquid phase. A galvanic DOS was used to measure the DOP and the results were fitted to the previous DO model. The parameters were fit and the fitting results indicated that using a new two-phase friction model did not significantly improve the fitting parameters when compared to using the old Fanning friction model. However, since the newer friction model fit the pressure profile better, for the sake of consistency and fidelity, the new friction model is still recommended when prediction bubble size for MBE.

Finally, the knowledge and conclusion obtained during the mass-transfer experiment was used to simulate the performance of an industrial-scale reactor aerated by MBE nozzle formations. The final simulation results can be used to help engineers and scientists to design reactor vessels with high gas-transfer demand and improve the production rate of two-phae reactions.

An unexpected conclusion reached was that the lab-made 7 mm MBE performance is on par with the market-ready Riverforest commercial 7 mm model.

### **FUTURE WORKS**

During the scale-up process for ejector-sparged industrial-scale reactor, both the mass transfer property and the geometry of the mass transfer unit cell needed to be characterized. The mass transfer property of microbubble ejector was characterized using the steady state dissolved oxygen experiment. However, the geometry of the mass transfer unit cell was not investigated thoroughly to observe the effect of gas fraction on the cell geometry.

The existing literature that was used to derive Eqn 6-3 was based on liquid-liquid entrainment. The 12° angle used for Eqn 6-1 through 6-3 was measured when turbulent liquid jet was injected into liquid body. No gas fraction was present in the study that identified the 12° angle within the jet cone side profile. Given the importance of this angle on the geometry of the mass transfer unit cell, it is crucial to accurately measure the true angle from the jet cone centerline to the cone boundary.

Similar strategy used by Cushman could be applied to approach this investigation. A camera can be used to identify the boundary between the liquid jet and bulk fluid body with some image processing techniques such as increasing image contrast.

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