KWR 2021.115 | February 2022

Power to Protein - Fundamentals Hydrogen Transfer

Final report

TKI Water Technology

KWR

Bridging Science to Practice

Collaborating Partners

KWR

Avecom
Bioproducts & Apps

ALLIED WATERS®

TOPSECTOR
WATER &
MARITIEM

Final report

Power to Protein - Fundamentals Hydrogen Transfer

KWR 2021.115 | February 2022

Project number
402821

Project manager
Frank Oesterholt

Client
TKI Water Technology

Authors
Siddharth Seshan, Bas Wols and Johann Poinapen

Quality Assurance
Emile Cornelissen

Sent to
This is a public report.
This activity is co-financed with PPS-funding from the Topconsortia for Knowledge & Innovation (TKI's) of the Ministry of Economic Affairs and Climate.

Keywords
power to protein; hydrogen; mass transfer; modelling

Year of publishing
2022

More information
Johann Poinapen, *PhD, FIEAust, CPEng*
T +31 30 606 9638
E johann.poinapen@kwrwater.nl

PO Box 1072
3430 BB Nieuwegein
The Netherlands

T +31 (0)30 60 69 511
E info@kwrwater.nl
I www.kwrwater.nl

Summary

In this study, the aim was to gain fundamental insights into the mass transfer of hydrogen in bioreactors in order to promote the biosynthesis of single-cell protein. This is based on the findings from previous project phases (1 and 2) which involved feasibility and experimental studies conducted to progress the highly promising power to protein concept, as investigated by the companies Avecom (Impetus), KWR and Allied Waters. Specifically, the analysis presented in this study targeted the pilot-scale bubble column reactor operated by Avecom to further understand and test the power to protein concept. The activities conducted in order to achieve the above aim are as follows:

- Literature Review: A literature study was conducted to understand what types of reactors are found in literature related to hydrogen mass transfer coefficient and to assess which reactor configuration offers high mass transfer efficiency. Furthermore, based on the optimal reactor configuration, further information on the key operating parameters that are related to the mass transfer of hydrogen gas were identified. Focus was also given in understanding the effect of pressure, gas hold-up and mass transfer coefficient in bubble column reactors as defined by the partners in the previous phase of the project.
- Knowledge Exchange with Experts: A meeting and knowledge exchange session was organised with key experts in the relevant field in order to present the research activities that were conducted in this study. Recommendations from the expert session were taken into consideration and the key takeaway messages are provided in this report. They confirmed that the bubble column reactor provides the best and right configuration for the power to protein concept and advised to obtain additional monitoring data to optimise the modelling of the reactor performance.
- Modelling Study: In this activity, a CFD model was developed and calibrated using the available data to adequately represent the pilot-scale bubble column reactor that is currently being operated by Avecom to further understand and test the power-to-protein concept. Based on the findings from the literature review, a CFD model was developed incorporating the key design and operating parameters of the power-to-protein bioreactor. Various simulations and sensitivity analyses were conducted in order to determine the effects of these different key operating conditions on the mass transfer of hydrogen and gas hold-up to maximise the yield of bacteria in the bioreactor. Based on the results from the model simulations, recommendations for the optimal design of a scale-up bioreactor have been provided. Additionally, the limitations of the current model due to unavailability of necessary data have also been reported as well as recommendations to obtain the necessary data required to further calibrate the model thereby increase the prediction accuracy.

From the literature review conducted, it was established that bubble column and slurry bubble column reactors are suitable reactor types or configurations to achieve high performance in gas-to-liquid mass transfer and better gas hold-up. Such a conclusion was confirmed by experts whom were consulted in a knowledge exchange meeting. Additionally, key design and operating parameters were also identified from literature and expert consultation to develop a CFD model that simulated the performance of the bubble column reactor under varying design and operating conditions. From the modelling study, it was concluded that the most important parameters to be considered with respect to improving performance were bubble size, pressure and aspect ratio. These key findings from the study supported the methodology followed by Avecom, providing additional confidence to Avecom and the project partners on the pilot scale bubble column reactor currently in operation. Recommendations were also provided in what parameter values can be considered for the design and operations of a larger scale reactor, as envisioned to be pursued by Avecom and its project partners. Additionally, key recommendations regarding the monitoring of the operating parameters through the deployment of sensors were provided. With the availability of additional data, further calibration and validation of the current CFD model can be conducted to increase its prediction accuracy.

Contents

1 Introduction

1.1 Background

With the global population expected to cross 10 billion people by 2060 (UN, 2019), sustainable provision of food, energy and water continues to pose a challenge worldwide. As an effort to develop innovative and cross-cutting solutions to tackle such challenges, the recovery of resources from waste and used water streams has been a promising and prominent path forward. Furthermore, resource recovery is an essential contribution in achieving the goals set by the European Union to transition to a circular economy while being resource efficient, having a lower carbon footprint and at the same time maintaining a competitive economy. In this regard, the food industry is a sector that can greatly benefit from technological innovation to ensure a sustainable supply owing to the exponential increase in demand. The production of microbial protein from raw materials available in wastewater streams is one such technique that has the potential to provide solutions to the future global food challenges. Furthermore, the production of proteins from fast-growing microorganisms provides an alternative source to conventional protein sources from meat and fish that is deemed unsustainable and negatively contributing to climate change. Additionally, by providing the necessary energy from renewable energy sources, the three pillars of the water-food-energy nexus is further strengthened in one overarching innovative concept, which was thus named – Power-to-Protein (KWR 2018.078, 2018)

1.2 Power-to-Protein Concept

The Power-to-Protein concept was launched by Professor Willy Verstraete, University of Ghent (Belgium). Subsequently, the company Avecom, who is affiliated with University of Ghent and is led by Professor Willy Verstraete, developed the biosynthesis of single cell high-value protein (SCP through the utilisation of lithographic hydrogen-oxidising bacteria in a lab-scale reactor system (ref). Furthermore, Avecom is also researching the production of SCP by providing a part of the necessary raw materials from the organic compounds prevalent in wastewater. This concept is depicted below in Figure 1. For the production of the SCP, raw materials such as hydrogen acting as the biomass energy supply, ammonium (NH_4^+), carbon dioxide (CO_2) and oxygen are necessary. NH_4^+ is abundantly available in the reject water generated during the processing of sludge to produce biogas through anaerobic digestion within the wastewater treatment cycle.

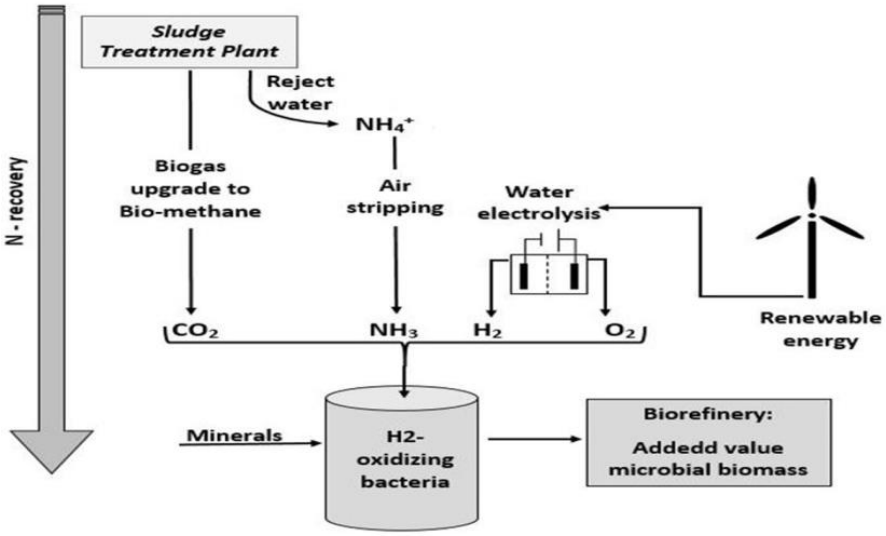

Figure 1: Power-to-Protein concept coupled to the wastewater cycle

Furthermore, during the upgrading of the biogas to bio-methane, CO_2 is released which can also be captured and used for SCP production. Finally, hydrogen (and oxygen) as needed by the hydrogen oxidising bacteria as an energy source, can be supplied via its production from the electrolysis of water. The energy supply needed from the electrolysis process can be further generated from renewable energy sources or by using surplus electricity during off-peak hours. Therefore, through such an innovative and cross-sectoral approach, a product of high demand of this protein can be produced or created from residual materials that are conventionally considered as waste products.

In 2014/15, during a Phase 1 study, an inventory was conducted within the TKI Water Technology (WT) framework to assess the viability of linking the Power-to-Protein concept with the wastewater chain. The idea was tested using the greater Amsterdam urban area as a case study (KWR 2015.049, 2015). The study revealed a great potential where 6,300 tons of protein can be produced annually while utilising the NH_4^+ available in the reject water separated from the digested sludge in the Amsterdam West WWTP. Such a quantity of produced protein can provide 36% of Amsterdam's population protein demand. A list of recommendations were also provided for further research which were partly pursued in a follow-up TKI WT Phase 2 study (KWR 2018.078, 2018). The scope of the study conducted in Phase 2 entailed the design of a pilot reactor with a target production capacity of 1 kg of protein per day while ensuring the safe and effective input of the raw materials into a suitable reactor configuration. Furthermore, the study included the investigation of the optimal reprocessing methods for the key raw materials such as ammonium and hydrogen given their greatest influence on the economic feasibility of the concept. Finally, the link between the ammonium from the wastewater chain and other raw materials such as hydrogen was meant to be made within the pilot study. From the Phase 2 study, the main conclusions were as follows:

- The targeted volumetric production capacity of the SCP was not achieved in the investigated pilot reactor. This was attributed to the insufficient mass transfer of hydrogen from the gaseous phase in the bubbles into the liquid phase which in turn lead to the reduced availability of hydrogen for the hydrogen oxidising bacteria. As a result, the pilot study in Phase 2 did not provide further insights needed to revise the economic evaluation that was conducted in Phase 1.
- The quality of the SCP was concluded to be inferior and was also attributed to the insufficient mass transfer of the gases, particularly hydrogen from the gaseous phase into the liquid.

Based on the results from the detailed study conducted in Phase 2, the following recommendations were listed thereby providing directions for future research:

- A literature review should be conducted to gain more fundamental understanding of the mass transfer of hydrogen and other gases in the bioreactor to promote the biosynthesis of the SCP.
- A modelling approach should be pursued to simulate different operating conditions that would improve the mass transfer of hydrogen and other gases.
- Through conducting a literature review and modelling study, options for a scalable and optimal reactor design while upholding adequate safety measures could be identified. With such an optimised reactor system, new pilot tests can be conducted to determine the economic evaluation for SCP production.
- In a well-functioning reactor system, further research should be conducted to understand the presence of thermoresistant bacteria in the microbiome when using recovered ammonium from the digestate (This was not in the scope of the current study).
- Further research should be conducted into the presence of hydrophobic persistent organic micro-pollutants (OMPs) in the ammonium sulphate in the digestate (This was not in the scope of the current study).
- Investigations should be carried out to identify the optimal procedure to reprocess the biomass into a dried protein product, while considering the protein quality (This was not in the scope of the current study).
- The economic feasibility of producing SCP using industrial ammonium should also be carried out (This was not in the scope of the current study).

1.3 Research Questions

Following partly the recommendations provided in the Phase 2 study, the following research questions were selected, investigated and reported in this report. These were selected because the most urgent factor that hampers the development of this power-to-protein concept is the insufficient mass transfer of hydrogen and is therefore a high priority.

- What is the most suitable reactor configuration to operate while considering various operating conditions and reactor geometry?
- Which operating parameters are crucial and heavily influence the mass transfer of hydrogen within a bioreactor?
- What are the important aspects to consider in scaling up the optimal reactor configuration?
- Can the most optimal reactor configuration be modelled using a Computational Fluid Dynamics (CFD) or mathematical model to determine:
    - The optimal reactor aspect ratio?
    - Optimal pressure?
    - Optimal gas holdup, gas recirculation and KLa?
    - Impact of bubble size?

1.4 Aim of the investigation

The overarching aim of this current study is to gain fundamental insights into the mass transfer of hydrogen in bioreactors to promote the biosynthesis of single-cell protein. The investigation should provide further insights for the design of a bioreactor where the input of cultural fluid, hydrogen and other gases is optimised and additionally gives sufficient confidence to further scale up the power-to-protein concept in a follow-up phase.

1.5 Activities

To answer the formulated research questions as listed in Section 1.3, the following research activities were conducted:

- Literature Review: Conduct a review to understand what types of reactors are found in the literature using hydrogen transfer and which specific type of reactor configuration is considered optimal. How is this reactor operated and what are the main points of attention? What can we learn from literature about gas hold-up, mass transfer coefficients in bubble columns? What specific information is available on mass transfer of hydrogen in bioreactors?

- Knowledge Exchange with Experts: Organise a meeting and knowledge exchange session with key experts in the relevant field in order to present the research activities conducted in this study and seek their advice and recommendations on the optimal reactor type, key operating parameters and reactor performance modelling.

- Modelling Study: Develop a mathematical or CFD model aiming at an optimal operation of the selected bioreactor regarding to H_2 transfer? Specific design and operating parameters of interest such as pressure, bubble size, reactor height, gas velocity, recirculation rates and bacterial growth will be included in the development of the model. The model will be calibrated using the available data from the power-to-protein pilot plant operated by Avecom to further understand the effects of these key operating parameters on the mass transfer of hydrogen.

2 Literature Review

2.1 Findings of Literature Review

A targeted literature review was conducted to understand what types of reactors are found in literature related to hydrogen mass transfer coefficient and the key design and operating parameters for optimal mass transfer efficiency. In this section, a discussion is presented on the findings of this literature review conducted to answer the research questions described in the above section.

2.1.1 Reactor Types in Literature

In different types of industries such as the biochemical, bioprocesses, chemical and petrochemical industries, varying bioreactor configurations are used for conducting different reactions and production of key resources. For example, aerated stirred tanks, particularly stirred dual impeller gas-liquid reactor, have been utilised and investigated to better understand its efficacy in the transfer of gases into the liquid phase to further stimulate the reaction or biosynthesis processes (Bouaifi et al., 2001). For the Fischer-Tropsch (FT) synthesis multiple reactor configurations are also used. FT process is a well-known process where indirect coal liquification or a conversion of a mixture carbon monoxide and hydrogen or water gas into liquid is conducted (Kantarci et al., 2005). Many reactors such as the multi-tubular fixed-bed reactors, circulating fluidised bed reactor and slurry reactors, primarily bubble columns, are commonly used for the FT process (Wang et al., 2007). With respect to methanol and dimethyl ether (DME) synthesis, slurry bubble column, slurry airlift reactors, fixed bed and fluidised bed reactors have been studied (Wang et al., 2007). Specific to gas-to-liquid processes, slurry reactors have been considered ideal and are most studied. When solids are present, thereby resulting in a liquid-solid suspension, such reactors are termed as slurry reactors (Kantarci et al., 2005; Wang et al., 2007). There are different types or configurations of slurry reactors including bubble columns, internal-loop airlift reactor, external-loop airlift reactor and spherical reactor (Wang et al., 2007). Specifically, bubble columns are cylindrical reactors in which one or many gases are bubbled through a liquid (Kantarci et al., 2005). The liquid can be fed into the reactor in either batch or continuous flow and the gas is injected through a gas distributor at the bottom of the vessel. While the airlift reactors and the spherical reactors have exhibited special advantages, bubble column reactors are easier to construct (Kantarci et al., 2005; Wang et al., 2007). Further advantages of bubble column reactors include low operational and maintenance costs due to no moving parts, excellent heat and mass transfer characteristics, ability to handle solids, and low pressure drops (Kantarci et al., 2005; Kumar et al., 2012; Wang et al., 2007).

Given the multiple advantages of bubble column reactors when compared to other reactors used for gas-to-liquid processes, such reactors are widely used in a range of practical process applications such as the Fischer-Tropsch (FT) synthesis, liquid phase methanol synthesis, biochemical processing, photo-bioreaction, wet-air oxidation, hydrogenation, oxidation and effluent treatment (Kantarci et al., 2005; Wang et al., 2007). As a result, bubble column reactors have proven to be a widely used reactor configuration in two- or three-phase systems, where the promotion of the mass transfer of the gas into the liquid or solid-liquid phase is achieved while maintaining an optimal gas hold-up. Furthermore, the use of other slurry reactor types specific to the mass-transfer of hydrogen is uncommon and in this respect, bubble column reactors have been investigated more. A diagram is presented that represents a clustering analysis that was conducted using text data (Figure 2). The analysis was conducted using the software VOSviewer (Version 1.6.17; van Eck & Waltman, 2021). Through this analysis, keywords mentioned within the titles and abstracts of a specialised literature search are inserted (Figure 2). The specialised literature search involved a SCOPUS search that included the following keywords: (KEY(bioreactor OR reactor) AND KEY(type OR configuration) AND KEY(hydrogen) AND KEY (mass-transfer)).

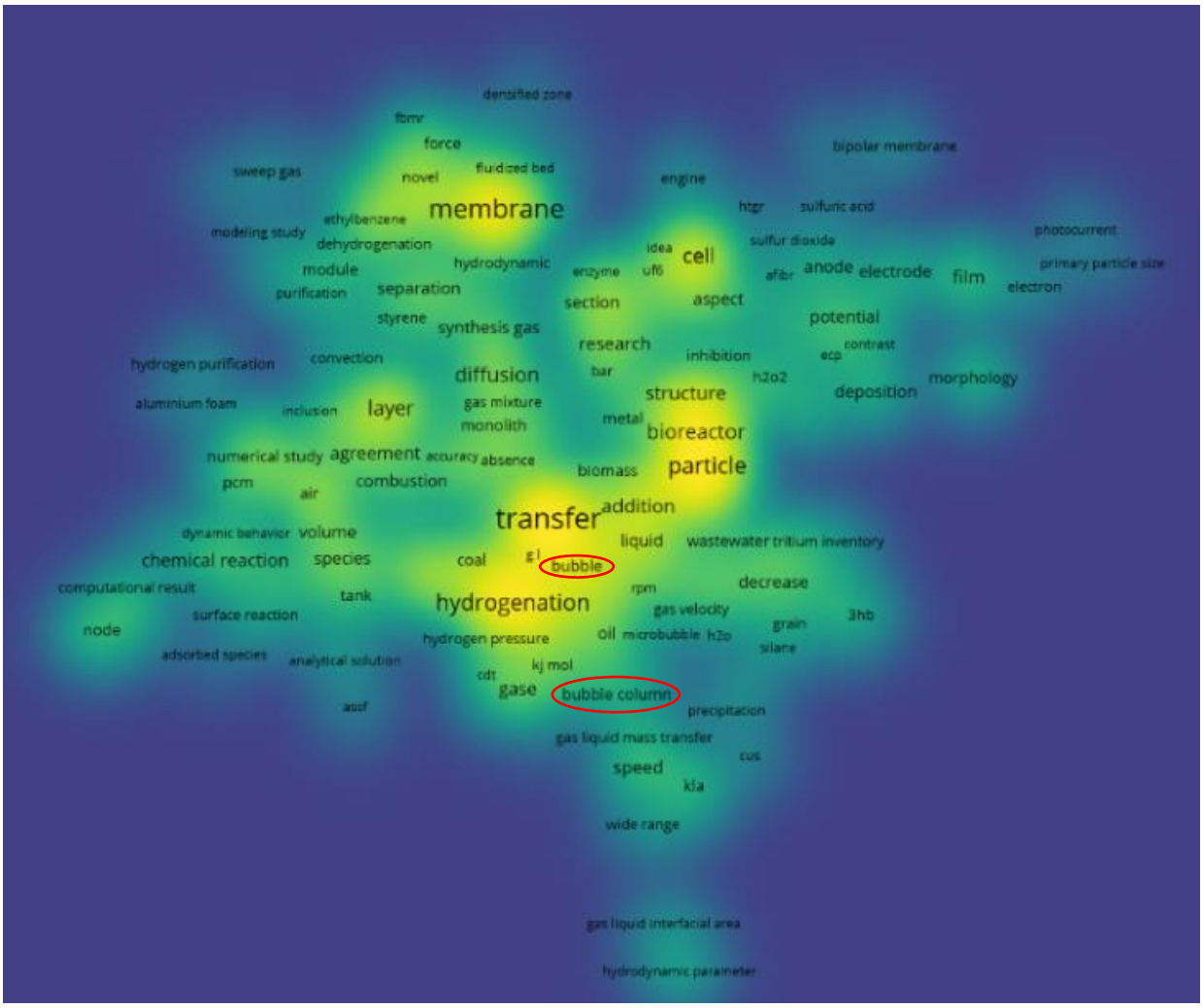

Figure 2: Density Visualisation of key clusters identified using a textual data as input from the Scopus search: (KEY(bioreactor OR reactor) AND KEY(type OR configuration) AND KEY(hydrogen) AND KEY (mass-transfer)). Keywords 'bubble' and 'bubble column 'are highlighted in red

The literature search targeted the type or configurations of (bio)reactors that are used in investigations related to the mass transfer of hydrogen. The analysis provides a visualisation where the prominent words mentioned most represent the centre of a cluster with the yellow colours surrounding the word representing the density of the cluster. In essence, the proximity of two words signify the co-mentioning of the words within the investigative studies. Therefore, the closer the words or the concepts are to one another, the higher the likelihood that those concepts have been investigated within the same study. While considering the *transfer* cluster density, it can be seen that *bubble* and *bubble column*, as highlighted in red, are close or in the proximity, respectively. Furthermore, keywords indicative to the use of hydrogen such as *hydrogenation* and *hydrogen pressure* are also within the same cluster density. Another concept of interest, *gas liquid mass transfer*, can also be seen to be found next to *bubble column*. In contrast, other potential reactor types of configurations such as a *membrane-based* bioreactor appear to be prominent in another spatial location within the cluster space. This indicates that with respect to the transfer of mass of hydrogen, from the gaseous phase into the liquid phase, bubble column reactors appear to be more widely investigated and implemented in the investigated literature.

Based on the above findings, it was concluded that bubble column or slurry bubble column reactors can be considered suitable reactor types or configurations with respect to achieving the high performance of gas-to-liquid mass transfer. Furthermore, specific to the mass transfer of hydrogen, bubble column reactors are more suitable. As

a result, the subsequent sections of the literature review provide the results of the findings specific to bubble column reactors.

2.1.2 Key Design and Operating Parameters for Bubble Column Reactors

As previously mentioned, bubble column reactors can be considered more advantageous compared to other reactors (such as the multi-tubular fixed-bed reactors and circulating fluidised bed reactors) both in design and operation. The main advantages of bubble column reactors are the efficient heat and mass transfer features, lower operating and maintenance costs due to very few moving parts and the ability of the reactors to easily handle solids. As a result, bubble columns are mostly used as multiphase reactors in chemical, petrochemical, biochemical and metallurgical industries (Degaleesan et al., 2001). In Figure 3 provided below, a bibliometric network was constructed and visualised using text data as input in the VOSviewer software (Version 1.6.17; van Eck & Waltman, 2021).. The specialised literature input for this analysis was to better understand which operational parameters are widely considered crucial and important to optimise in a bubble column bioreactor. The specialised literature search for this analysis also included a SCOPUS search with the following keywords: (KEY(bioreactor OR reactor) AND KEY(bubble column) AND (operation OR operate)) Additionally, the network provides insights on which operating parameters have been studied in detail to investigate how sensitive the performance of a bubble column reactor is to a given operating parameter.

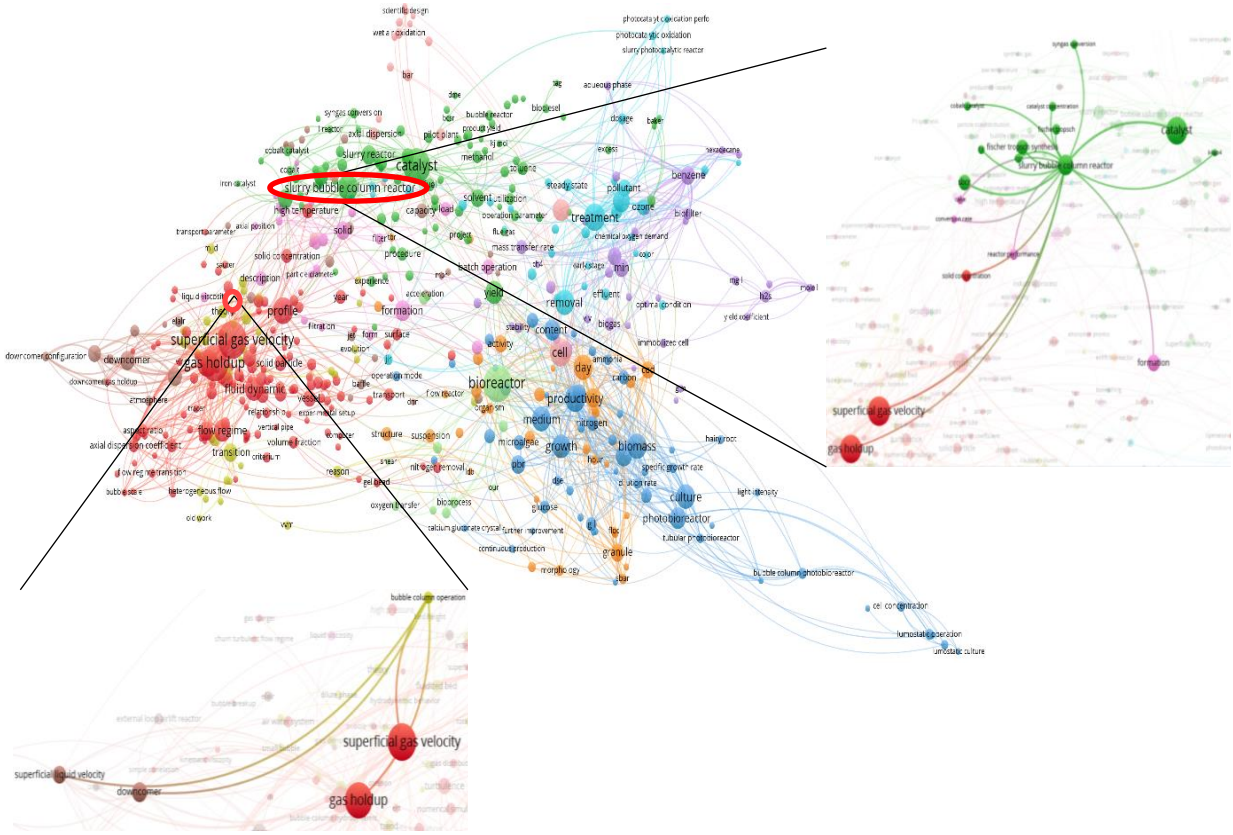

Figure 3: Network visualisation of co-occurence and inter-connections between prominent keywords using textual data as input from a Scopus search: (KEY(bioreactor OR reactor) AND KEY(bubble column) AND (operation OR operate)). The nodes 'bubble column operation' and 'slurry bubble column reactor' have been zoomed in.

Two prominent nodes in the network were further analysed, as depicted in Figure 3, within the zoomed in sub-figures. The nodes represent studies that have mentioned *bubble column operation* and *slurry bubble column reactor*. With respect to the *bubble column operation* node, links were found to exist with *superficial gas* velocity, *superficial liquid* velocity, gas *holdup* and *downcomer*. These links signify four relevant operational parameters of a bubble column,

which have been studied and concluded to be crucial with respect to operating bubble columns. Similarly for the *slurry bubble column reactor* node, more operating parameters significant to its operations have been linked. The key ones identified were *superficial gas velocity*, *gas holdup*, *catalyst*, *catalyst concentration*, and *solids concentration*.

Given the intention of increasing the mass transfer within a bubble column through the increase in gas holdup, literature studies have also confirmed and complemented the results from the bibliometric network analysis visualised and detailed above. Specifically, the following important variables were identified that affect the gas holdup, bubble dynamics and flow regime in a bubble column (Deckwer & Schumpe, 1993; Li & Prakash, 1997; Luo et al., 1999; Shah et al., 1982):

- Static height and column diameter
- Pressure
- Superficial gas velocity
- Liquid velocity
- Solids concentration
- Design of gas distributor

2.1.3 Influence of key operating parameters on the gas holdup and mass transfer in a bubble column reactor

The gas holdup in a bioreactor can be defined as the volume of gas dispersed from the gaseous phase into the liquid phase. In the following sub-section, a review of literature was conducted on the specific operating parameters listed in Section 2.1.2 above. For each operating parameter, its influence on the gas holdup and mass transfer of a bubble column reactor has been discussed.

(a) Static height and column diameter

In Kumar et al. (2012), experiments were performed to measure the effect of the static liquid height on the overall gas holdup for a bubble column. Additionally, pressure drops were measured to calculate the overall gas holdup and the experiments were carried out for a 1.0 and 1.8 m static liquid height. It was concluded that for the investigated static heights of liquid, minimal effect on the gas holdup was observed. Similarly, in the experiments conducted by Sasaki et al. (2015), the effect of the initial liquid height on the total gas holdup for an air-water bubble column was investigated. The results of this study corroborated the initial findings that the influence of differing liquid heights on the gas holdup is minimal, as also can be depicted from the results provided below in Figure 4. It was also concluded that the flow regime mainly depended on the superficial gas velocity and the influence of the height is small. Furthermore, a correlation was found between the gas holdup at varying liquid heights with the Froude number, which is computed by using the superficial gas velocity and the initial liquid height.

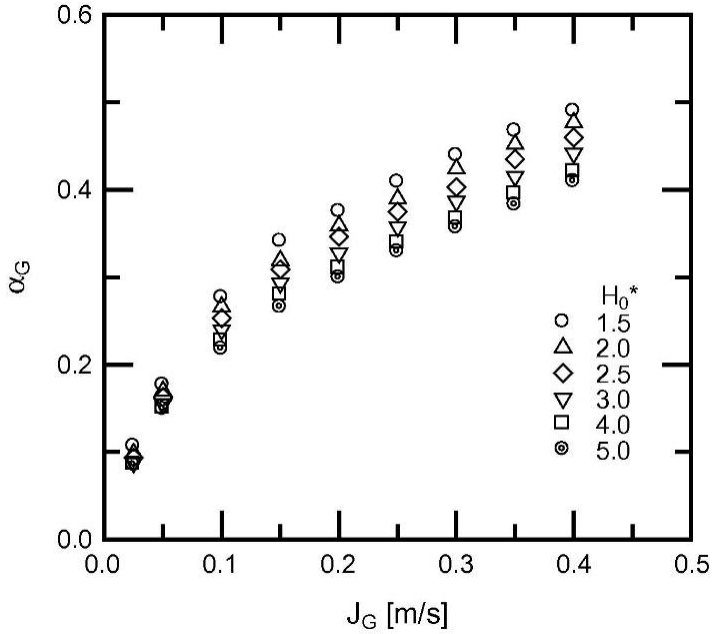

Figure 4: Results from an experimental study comparing different initial liquid water heights at differing superficial gas velocity. Taken from Sasaki et al. (2015).

Daly et al. (1992) studied through experiments the effect of column diameter on the gas holdup. It was observed that at a gas velocity greater than 0.08 m/s, the Sauter mean bubble diameter, as defined as the average size or diameter of the bubbles in the column, remained unchanged in larger diameter columns. In contrast, for smaller diameter columns the Sauter mean bubble diameter increases with increasing gas velocity. This was attributed to the presence of larger bubbles. Generally, it was noticed that in smaller diameter columns, the Sauter mean bubble diameters were higher. The difference was attributed to the different flow regimes that become prominent for the differing column diameters. At the gas velocities investigated, the large diameter columns display a churn-turbulent flow regime, which leads to the increase in liquid circulation and turbulence. This results in the formation of smaller bubble columns, and thereby, a higher gas holdup. For smaller column diameters, a slug flow regime is witnessed, thereby leading to the presence of larger bubbles, and subsequently lower gas holdups. To summarise the influence of static liquid height and column diameter, literature shows that an aspect ratio, that is the ratio of height and the diameter of column, greater than 5 and a diameter greater than 10-15 cm; the effect of liquid height over gas holdup is negligible (Luo et al., 1999; Shah et al., 1982).

(b) Pressure

Literature studies have predominantly concluded that the gas holdup increases with the increase in pressure. At higher pressure, higher gas densities is expected which subsequentially leads to lower bubble rise velocities. This ensures a larger residence time of the gas bubbles. In the study conducted by Letzel et al. (1999), the difference in gas holdup at elevated pressures compared with atmospheric pressure was investigated. The results from the study is illustrated in Figure 5. In the study, a 0.15 m diameter bubble column with a height of 1.22 m was used. Nitrogen gas was injected using a perforated plate sparger into the liquid phase (water). The pressure of the system was varied up to 1.3 MPa. As evident in Figure 5, with the increase in pressure in the bubble column, an increase in gas holdup was observed.

In similar published studies, it was concluded that with the increase in pressure, bubble diameter can decrease as compared with the size of the bubble initially formed due to the gas distributor openings (Idogawa et al., 1985). Furthermore, under normal pressure conditions, a variety of bubble sizes and wider range of diameter distribution was observed. Whereas, at higher pressures, a narrower bubble size distribution or more uniform bubble sizes are seen (Idogawa et al., 1985). Similarly, in a study conducted by Kang et al. (1999), bubble properties in a 0.058 m

diameter and 1.5 m height bubble column was studied under pressure fluctuations, with the highest pressure incorporated being 0.5MPa. The results of such a study confirmed that the bubble size and rising velocity does decrease with an increase in pressure, with the bubble size distribution becoming narrower. It was also deduced that the higher pressure, as a result, leads to a homogenous bubble flow regime even at relatively higher gas velocities.

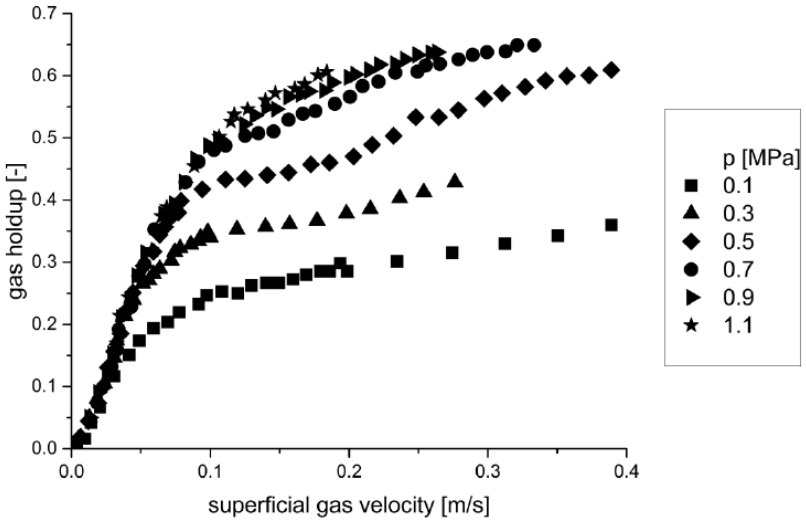

Figure 5: Effect of increasing pressure in a bubble column to the gas holdup. Taken from Letzel et al. (1999)

(c) Superficial gas velocity

In general, published studies in literature have shown that increasing the superficial gas velocity leads to an increase in the gas holdup (Daly et al., 1992; Krishna et al., 1997; Saxena et al., 1990). Furthermore, the dependence of the gas holdup on the superficial gas velocity has been defined by the following power-law expression (Deckwer, 1991):

$$\varepsilon_g = k * U_g^n$$

where ε_g Is the gas holdup (unitless); Ug in m/s is the superficial gas velocity; k is a constant (s/m); n is dependent on the flow regime.

Generally, at low superficial gas velocities, bubbles are small and uniform in size (Hyndman et al., 1997). On the other hand, at high superficial gas velocities, the coalescence rate of the bubble increases significantly, and therefore, the gas-liquid flow becomes heterogenous within the bubble column containing a mixture of small and large bubbles (Schumpe & Grund, 1986). In the experiments conducted by Kumar et al. (2012), the effect of superficial gas velocity on gas holdup was also investigated. In Figure 6 below, the results from the experiment have been illustrated where it can be seen that there is a sharp increase in the gas holdup, almost linearly, which increased with superficial gas velocity. At higher superficial gas velocities, the slope decreases slightly. These two different slopes correspond to the homogenous and heterogenous flow regime, respectively.

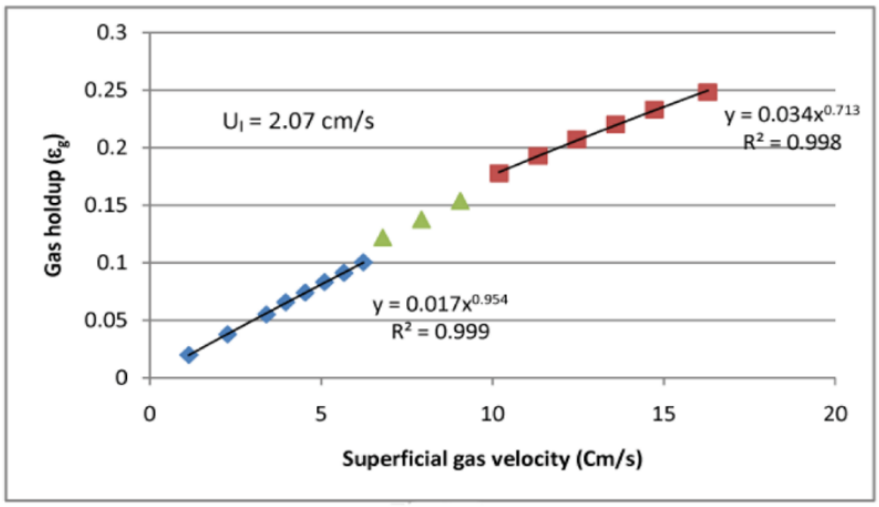

Figure 6: Effect of superficial gas velocity on the gas holdup of a bubble column. Taken from Kumar et al. (2012)

(d) Liquid Velocity

With respect to the liquid velocity, studies conducted showed that the gas holdup decreased with an increase in liquid velocity. In the experiments conducted by Hills (1976), the gas holdup was measured in a 15 cm diameter bubble column at superficial gas velocities of 0.07 – 3.5 m/s and a liquid velocity of 0.0 – 2.7 m/s. The results of the experiment suggested a decrease in the gas holdup with an increase in the liquid velocity. Similarly, Fujie et al. (1979) and Friedel et al. (1980), reported a decrease in the gas holdup with an increase in liquid velocity in downflow bubble columns of 45 cm and 15 cm internal diameter respectively.

In the experiments conducted in Kumar et al. (2012), varying liquid velocities up to 16.04 cm/s in co-current flow, were evaluated for its influence on the gas holdup. Furthermore, the effect of liquid velocity was studied for two phase flows as well as three phase flows, under different solid concentration in the slurry. With respect to two phase flow, it was observed that the overall gas holdup decreased with increasing liquid velocity, as shown in Figure 7. Specifically, the effect of liquid velocity on gas holdup was seen to be less pronounced at low superficial gas velocities. With the increase in superficial gas velocities, the gas holdup decreased at a faster rate for change in the liquid velocity. Furthermore, co-current flow of both gas and liquid results in an increase of the bubble rise velocity, thereby resulting in the bubbles leaving the column faster or a decrease in the residence time of the gas phase. This in turn also results in a lower gas holdup.

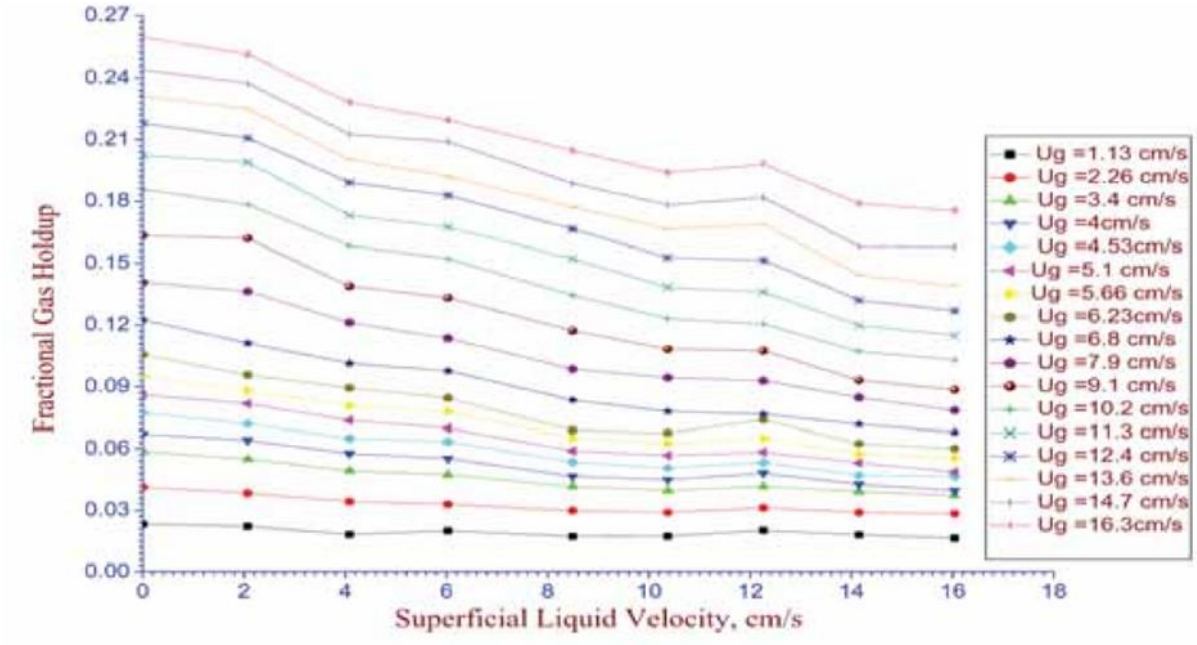

Figure 7: Effect of liquid velocity on the gas holdup for two phase co-current flow. Taken from Kumar et al. (2012)

With respect to the three phase co-current flow, the experiments were performed with different solids concentration up to 9% wt/vol. As shown in Figure 8, the solids loading of below 1% results in trends similar to a two-phase flow system. While at higher solids loading, such as the results shown in Figure 9 with 9% solids loading, the effect of liquid velocity over gas holdup is insignificant. High concentration of solid particles promotes the coalescence of bubbles, and thus decreasing the gas holdup.

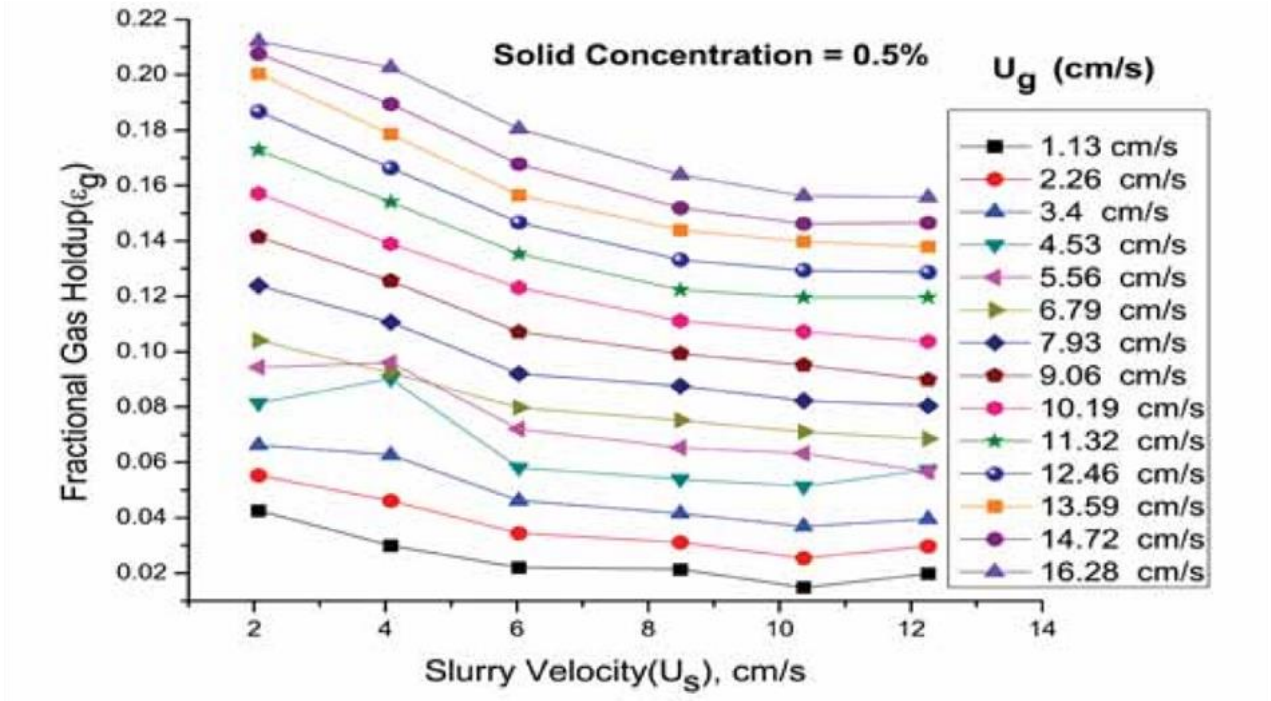

Figure 8: Effect of slurry velocity on the gas holdup for three phase co-current flow with a solids concentration of 0.5% wt/vol. Taken from Kumar et al. (2012)

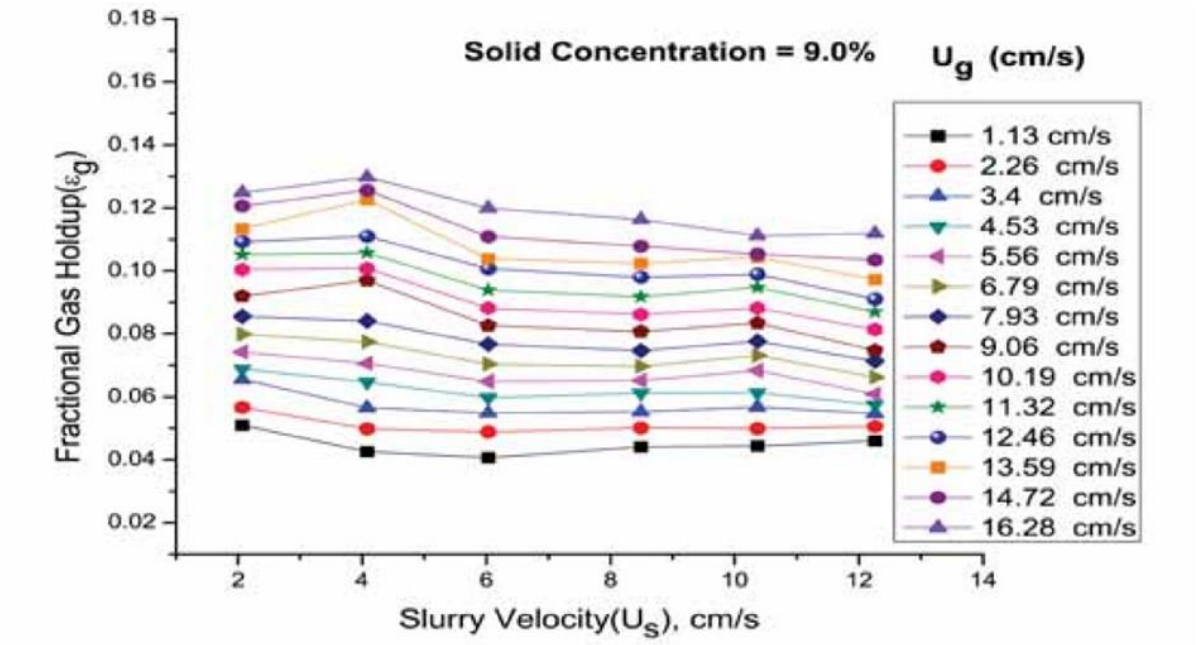

Figure 9: Effect of slurry velocity on the gas holdup for a three phase co-current flow with a solids concentration of 9% wt/vol. Taken from Kumar et al. (2012).

(e) Solids concentration

The influence of solids concentration has been reported in literature to decrease the gas holdup with increasing solids concentration (Fan et al., 1999), as also depicted in Figures 8 and 9 . It was also reported that the presence of solids increases the bubble size, resulting in fast bubbles (Su & Heindel, 2003). Moreover, with the reduction in the bubble breakup (i.e. increase in bubble size) and an increase in the mixture (liquid-solid) velocity, a decrease in the gas holdup was observed (Luo et al., 1997; Tsuchiya et al., 1997). The effect of slurry velocity with varying solids concentration on the gas holdup has been discussed in detail in the previous Section.

In the study conducted by Mena et al. (2005), experiments were conducted to study the effect of the presence of the solid phase with particle size 2.1 mm and density of 1023 kg/m^3 in a batch bubble column reactor with 0.14 m diameter. With the addition of the solid, the density and viscosity of the liquid change. Each particle surface creates a no-slip condition for the liquid, where the liquid velocity is zero. Therefore, extra velocity gradients are created and the viscous dissipation increases, leading to an increase in the viscosity and a decrease in the rise velocity of the bubble due to the increase slurry viscosity. Furthermore, with the hydrodynamic forces and the collision of bubbles and particles, the speed of the bubble reduces, leading to a reduction in the bubble rise velocity that increases the gas holdup. However, in viscous media, bubble coalescence is promoted, thereby resulting in the formation of bigger bubbles and the decrease in the overall gas holdup (Kumar et al., 2012). Such an effect is seen to increase with the increase in slurry concentration. Given the two competing mechanisms, a maximum in the gas holdup based on solids loading was achieved at 3-5% wt/vol (Kumar et al., 2012). In their experimental study, Kumar et al. (2012) used glass beads with 35 µm average size as solid particles in a three phase co-current air-water system. As discussed earlier, from the same experiment, the liquid velocity was varied and the solids concentration was also varied in two different ranges, from 0% to 1% (Figure 9) and from 1% to 9% (Figure 10). For all the liquid velocities, it was observed that the gas holdup was either constant or slightly increased up till a solids loading of 5%. Whereas for solids concentration above 5%, a depicted in Figure 10, a substantial decrease in the holdup was seen for solids concentration of 7% and 9%. In Figure 10, an optimal solids concentration of 0.7% can be concluded for a slurry velocity of 12.26 cm/s, and in Figure 11, an optimal solids concentration of 3% was seen for a lower slurry velocity of 6.03 cm/s .

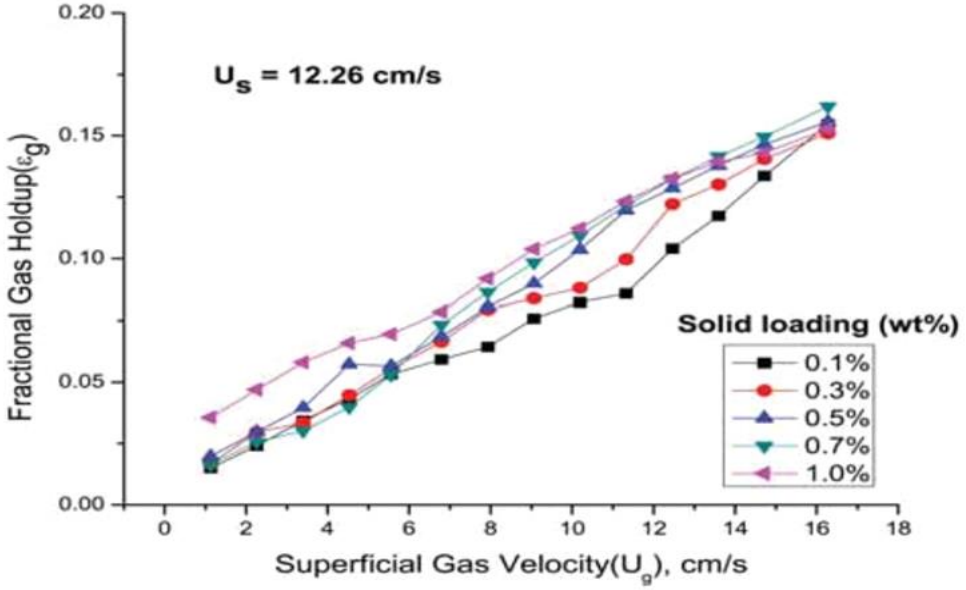

Figure 10: Effect of solids loading to the gas hold-up ranging from 0-1% wt/vol. Taken from Kumar et al. (2012)

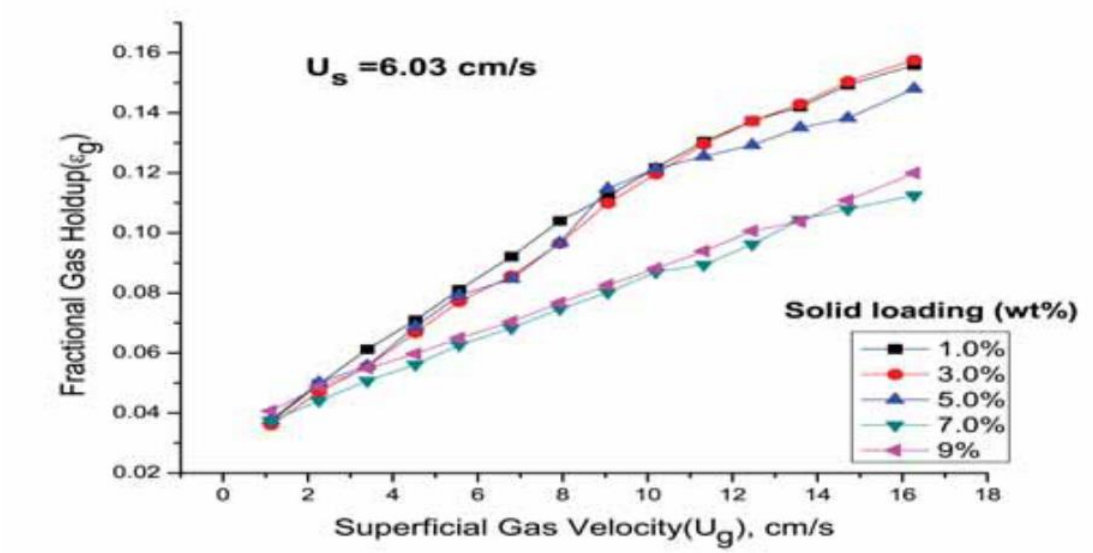

Figure 11: Effect of solids loading to the gas holdup ranging from 1-9% wt/vol. Taken from Kumar et al. (2012)

(f) Design of gas distributor

Over and above the column diameter and the properties of the liquid, the design of a gas distributor directly influences the properties of the bubbles injected into a bubble column reactor (Kumar et al., 2012; Luo et al., 1999). The design of a gas distributor includes the principle or method that the gas sparger is based on, diameter of the sparger holes, distance between the sparger holes and position of the distributor in the bubble column reactor. As a result, the type and design of a gas sparger determine the bubble characteristics which subsequentially leads to changes in the gas holdup. More importantly, the type of sparger used determines the bubble size that is observed in a bubble column (Kantarci et al., 2005). . In practice, there are many gas sparger types, namely – perforated plat, porous plate, membrane, ring type distributors and arm spargers (Kantarci et al., 2005). Literature also suggests that the smaller the bubbles, the greater the gas holdup values achieved (Bouaifi et al., 2001). Therefore, gas distributors that have smaller hole or orifice diameters tend to result in a higher gas holdup Furthermore, Schumpe & Grund (1986) in their experiments used a perforated plate and ring type gas distributor and concluded that the ring type distributor leads to a smaller gas holdup. Additionally, it was concluded that perforated plates contributed more to small and large bubbles compared to the ring type distributor.

3 Modelling of the Pilot-Scale Bubble Column Reactor

3.1 Literature to Modelling - Key Design and Operating Parameters

While considering the findings of the literature review on the key design and operating parameters with respect to bubble column reactors, a translation was conducted to incorporate the parameters into the modelling activities. Given below in Table 1, is a comprehensive overview of the key operating parameters found in literature as well as those based on the design and operating conditions of the pilot-scale reactor, operated by Avecom.

3.2 Current Power to Protein Pilot-Scale Bubble Column Reactor

Currently, Avecom is operating a pilot-scale bubble column reactor to investigate the implementation of the power to protein concept. The pilot-scale bubble column reactor is shown in Figure 12. At the bottom of the reactor, a sparger plate is installed, where a combination of gases including H_2, CO_2 and O_2, are fed at a specific flowrate. At the top of the reactor column, the mass of gas that has not transferred into the liquid is captured and recirculated to the bottom of the column at a given recirculation rate. Therefore, a combination of new and recirculated gases are fed into the column. The pilot reactor is operated at a specific overpressure and the reactor is run in batch mode with a duration between 5-8 days. Currently, continuous operations is also being investigated. The target values of the key performance indicators are provided in Table 2 along with the values achieved by the pilot-scale bubble column reactor. The detailed design and operational data of the pilot-scale reactor was provided by Avecom and was used in the CFD modelling as part of this project, but is subject to confidentiality.

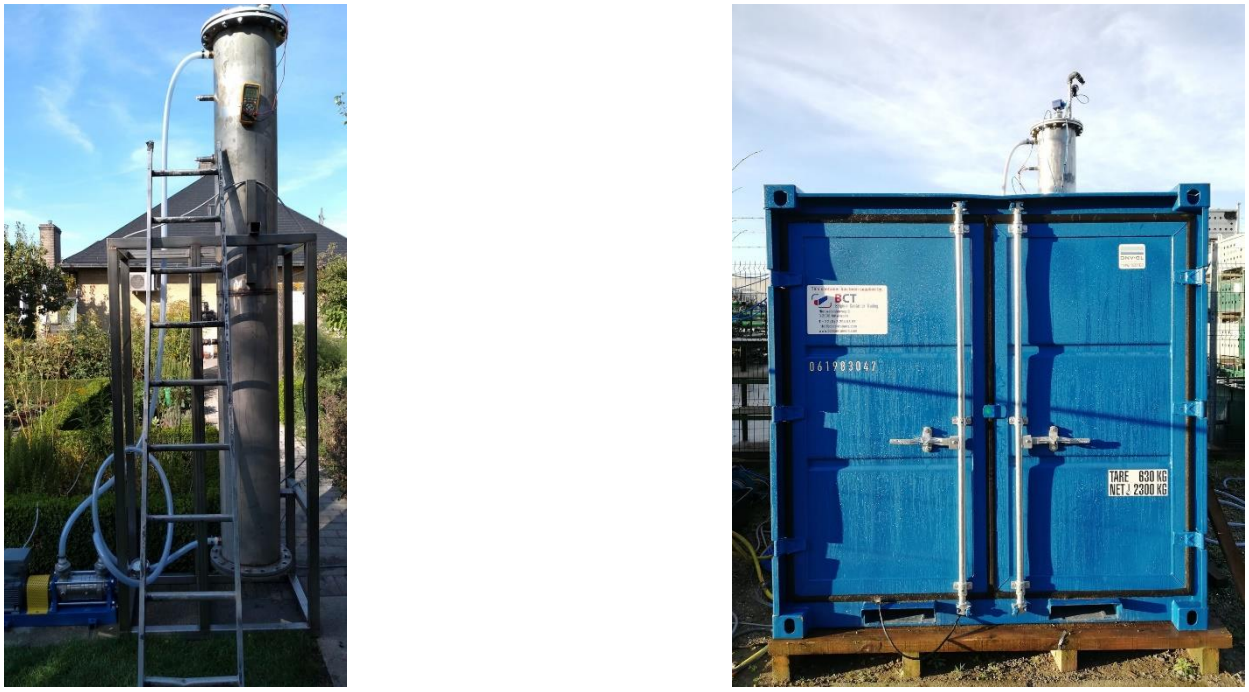

Figure 12: Current Power-to-Protein pilot-scale bubble column reactor, operated by Avecom

Table 1: Translating literature review findings to modelling activities. Key design and operating parameters have been listed and their inclusion in the model have been stated. Additionally, information has been provided on whether a sensitivity analysis was conducted for a specific operating parameter.

Operating Parameters Important for gas holdup in bubble columns as per Literature	Considered in model (Yes/No)	How has it been inputted/calculated in the model?	Sensitivity Analysis Conducted
Superficial Gas Velocity	Yes	Inputted based on pilot-scale data	No
Liquid Velocity	Yes	Inputted based on pilot-scale data	No
Bubble size	Yes	Inputted based on the size of the sparger holes	Yes
Liquid viscosity	Yes	Value for water used	No
Density of gas	Yes	Calculated from molar mass distribution in gas (H_2, CO_2, O_2)	No
Mass transfer coefficient H2 gas to liquid	Yes	Value calculated from empirical relation given by Yang et al. (2000)	Yes
Diffusion coefficient H2 in water	Yes	Inputted from literature data (DOI: 10.1615/AtoZ.d.diffusion_coefficient, 4.5E-9 m2/s)	No
Henry's coefficient H2	Yes	Inputted from literature data (Wikipedia, kH=1300 L.atm/mol)	No
Design of gas distributor	Yes	Two ways – (i) modelled as a constant boundary condition; (ii) Individual holes of sparger modelled	No
Solids concentration	No	Data on suspended solids to be provided by Avecom	N/A
Column diameter	Yes	Inputted when designing the column profile in the model. Based on design of the pilot.	Yes
Static water height (column height)	Yes	Inputted when designing the column profile in the model. Based on design of the pilot.	Yes
Overpressure	Yes	Inputted based on pilot-scale data	Yes
Bacteria growth parameters (taken from https://www.biorxiv.org/content/10.1101/847939v1.full.pdf)			
Yield hydrogen to bacteria growth	Yes	0.2 [g bact/g H2]	No
Maximum growth rate	Yes	3 [g/L/d]	No
Maximum bacteria concentration	Yes	8 [g/L]	No
Saturation constant	Yes	1E-2	No
Initial concentration of bacteria	Yes	0.1 [g/L]	No

Table 2: Key performance indicators with target values and performance of the pilot-scale bubble column reactor

Parameter	Target Value	Pilot-scale Bubble Column Reactor
Productivity (g CDW/L. d)	3 – 5	1.74
Yield (g CDW/ g COD H2-used)	0.2	0.17
Protein content (%)	>65	80

3.3 Knowledge Exchange with Experts – TU Eindhoven

On 30 August 2021, a knowledge exchange session was organised between the project team KWR Water Research Institute and experts from Technical University Eindhoven (TUE) in order to seek their advice on our approach to answer the research questions and to seek their recommendations on the optimal reactor type, key operating parameters and reactor performance modelling.

The experts were as follows:

- Dr. Ivo Roghair, Assistant Professor, Chemical Process Intensification, Department of Chemical Engineering and Chemistry.
- Dr. Maike Baltussen, Assistant Professor, Multi-scale Modelling of Multi-phase Flows, Department of Chemical Engineering and Chemistry.

The knowledge exchange proved to be a very valuable session where the experts from TUE expressed their interest in the work being conducted and are eager to learn on the outcomes of the project. The key takeaways from the discussion are listed below:

- The experts believed that the use of bubble column reactors is suitable for the application of ensuring higher mass-transfer and biosynthesis of single-cell protein. This provided further validation from academics in this field of research as already concluded from the Literature Review.
- The methodology used for the CFD modelling was deemed scientifically sound and the experts were in agreement with the process. They mentioned that the design and operating parameters affecting mass transfer of hydrogen, such as height, pressure and bubble size, are important to model. Further details on the methodology of the model development and simulations can be found in Section 3.4 below.
- The experts commented on the need for additional data through sensor deployment to further validation the CFD model. For example, bubble size is very important for gas holdup and mass-transfer of gases. Therefore, there is a need to collect data on such key parameters using cutting edge sensor and probe technologies available in the market.
- The experts posed an open-ended question regarding the output of the model. They suggested exploring modelling the protein production directly, instead of the bacteria concentration in the slurry. They themselves have never done that and are willing to explore this together if there is a next phase of the modelling exercise.
- Based on the experts' interest in the project and on the future outcomes, there is scope for collaboration to further investigate more fundamental questions with a goal to solve complex operational challenges faced in the current or subsequent pilot-scale studies.

3.4 Methodology and Development of the CFD Model

The methodology of the power-to-protein CFD model is described below. The model was developed in COMSOL Multiphysics 5.6.0.280 (COMSOL).

3.4.1 Physics

- Multiphase flow: the study of multiphase flows can be done at several scales. The physical scales may range from micrometres up to metres. Resolving all length scales is computationally impossible. For larger scale systems (like the bubble column we consider), it is not possible to track individual bubbles and study the gas-liquid interface. The presence of the gas and liquid phases are therefore modelled as volume fractions (dispersed multiphase flows, where the interfacial effects between bubble and liquid are treated as sources). The most inexpensive (with respect to calculation time) dispersed multiphase flow model is the bubbly flow model. In this model, the gas phase is the dispersed phase and the relative bubble velocity is described with an equation that balances the drag and pressure gradient. For the drag coefficient, a model for small spherical bubbles (Hadamard (1911); Rybczynski (1911)) was used. Also, the momentum balance is defined for the continuous phase. This approach is valid if gas hold-up remains small (<10%). In our system the gas hold-up is around 10%. The momentum balance is solved by the Navier-Stokes equations, where the turbulence closure is done by means of a standard k-ε turbulence model, which is an industry standard for CFD.

- Transport and reaction of (dissolved) hydrogen and bacteria. The mass transfer from hydrogen gas to the water phase (m_{gl} [g/s]) is calculated by means of the two-film theory as follows:

$$m_{gl} = k(c^* - c).M.a$$

  Where *k* is the mass transfer coefficient (m/s) from gas to water, *M* the molecular weight, *a* the surface area of the bubbles (calculated by the model) and *c** is calculated from Henry's law, where $c^* = \frac{p*}{H}$, where *p** is the partial pressure (as the molar concentration ratio of hydrogen in the gas is only about 11%) and *H* is Henry's coefficient. The partial pressure is calculated by the reference pressure plus the actual pressure calculated in the model divided by the molar mass ratio of hydrogen. The mass transfer is used as a source term in the transport modelling for dissolved hydrogen.

  The dissolved hydrogen (*S*) and bacteria (*X*) are transported by means of advection and diffusion. Reaction (bacterial growth) is modelled by a logistic growth model with Monod kinetics (Xu, 2020):

$$\frac{dX}{dt} = \mu X$$

$$\frac{dS}{dt} = -\mu \frac{X}{Y_{XS}}$$

$$\mu = \mu_{max}\left(1 - \frac{X}{X_{max}}\right)\left(\frac{S}{K_S + S}\right)$$

  where μ_{max} is the maximum growth rate (1/s), X_{max} the maximum concentration of the bacteria, and K_s the saturation constant. The rates *dX/dt* and *dS/dt* are added as sources and sinks in the transport modelling in COMSOL.

The combined gases that are fed to the reactor consists of hydrogen, oxygen and carbon dioxide at a specific operated ratio. Accounting for the molar mass of hydrogen (2 g/mol), oxygen (32 g/mol) and CO2 (44 g/mol), the molar mass ratio is 11.5% for hydrogen. Using these mass ratios, the gas density is equal to 0.507 g/L.

An overview of the various relevant constants used in the CFD model for the power-to-protein bioreactor is shown below in Table 3.

Table 3: 3 Overview of constants used in the modelling for the Avecom pilot installation

Constants	Value*
Bubble diameter	1 [mm]
Reference pressure	1.3 [bar]
Molecular weight of hydrogen	2 [g/mol]
Density of inlet gas	0.507 [kg/m3]
Mass transfer coefficient	6.7E-4 m/s
Henry's coefficient for H2	1320 L.atm/mol
Diffusion coefficient for H2	5.1E-9 m2/s
Monod constant	1 mol/m3
Yield hydrogen to bacteria (Yxs)	3.2 g-bact/mol H2
Maximum growth rate bacteria	3.472E-4 1/s
Maximum bacteria concentration	8 [mol/L]
Initial bacteria concentration	0.1 [g/L]

*The values for the constants were taken from various sources in literature.

The molecular weight of a bacteria is set to 1 g/mol, this was needed as the model works with molar concentrations, but it does not have influence on the model results when calculating back to g/L. The maximum growth rate is calculated from a measured number of 3 g/L/day [given by Avecom, more details can be found in Matassa et al. (2016)], divided by the initial bacteria concentration to get the maximum growth rate per second (s). The initial bacteria concentration was 0.1 g/L.

The mass transfer coefficient (k_L in [m/s]) is calculated from Yang et al. (2000):

$$k_L = 154.6\frac{D_{H2}}{d_b}.Eu^{0.0052}.Re^{0.076}.Sc^{-0.133}\pi r^2$$

Where D_{H2} is the diffusivity of hydrogen in water (5.1 E-9 at 25 C), d_b the bubble size diameter, *Eu* the Euler number ($Eu = \frac{P}{\rho_L d_b v^2}$, where *v* is the bubble rise velocity (estimated at about 0.4 m/s)), *Re* the Reynolds number ($Re = \frac{d_b v}{\eta}$, where η is the kinematic viscosity of water (1E-6 m^2/s)) and *Sc* the Schmidt number ($Sc = \frac{D_{H2}}{\eta}$).

3.4.2 Domain/mesh

The reference system consists of a cylinder with the dimensions as present in the pilot bubble column reactor. The geometry is schematised as a 2D axisymmetric system. The middle axis acted as a symmetry line, so that half of the geometry could be considered (that could be revolved around the central axis). A rectangular mesh with a mesh size of 0.5cmx0.5cm was used. Close to the side walls, 8 rectangular boundary layers were used to refine the mesh. The complete mesh consists of 19532 domain elements and 1104 boundary elements. A schematic diagram of the system and the mesh is shown in Figure 13.

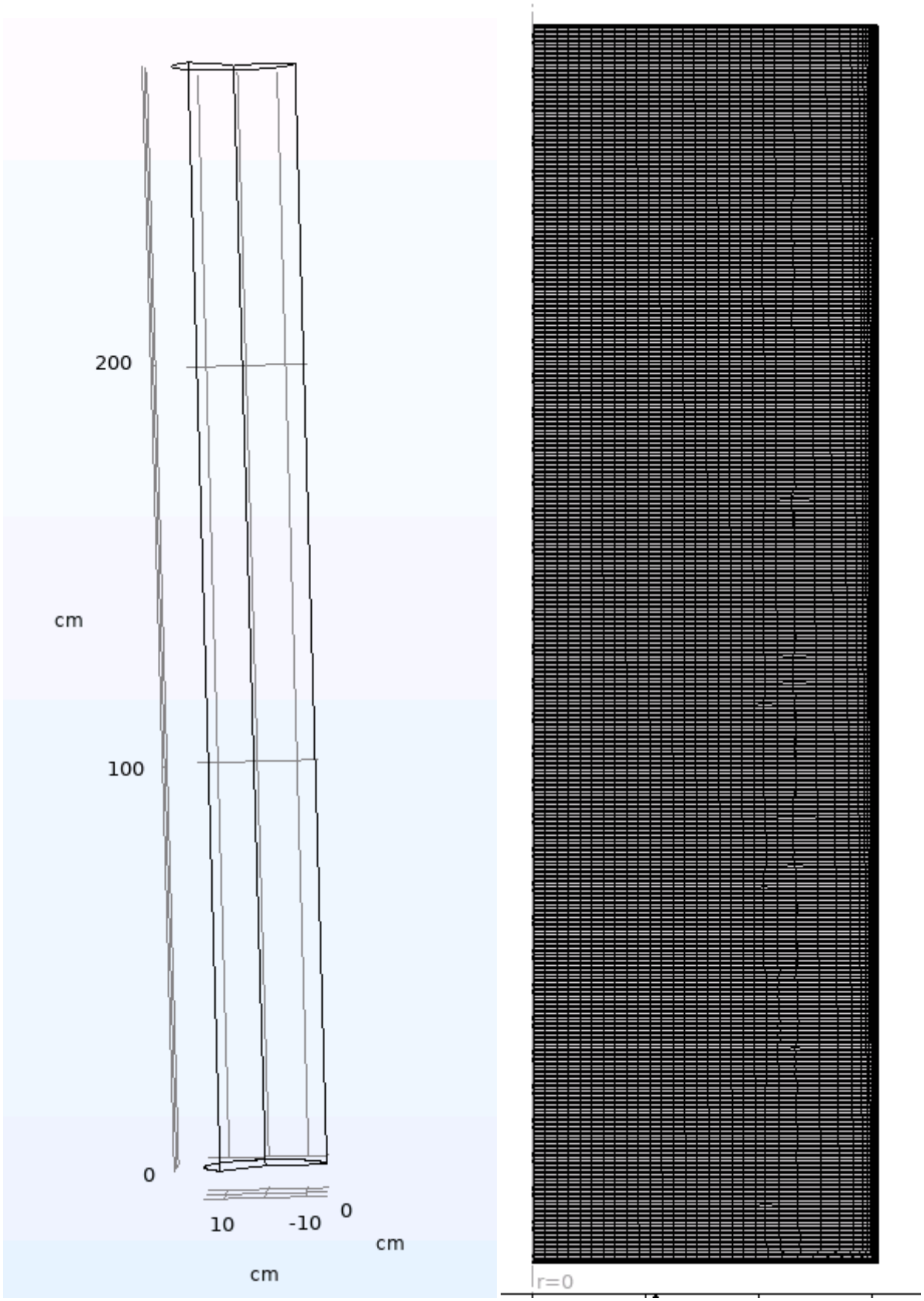

Figure 13 Geometry of the bubble column (axisymmetric system on the left) and rectangular mesh (on the right). Note that the axis are not equidistant to allow for a better visualization.

The gas inlet consists of a sparger with small holes (see Figure 14). The gas inlet was modelled in two different ways:

- Modelling the separate holes, however, the holes in the model could not be made as small as in the sparger, because the local gas hold-up became too high and the model became unstable.
- Modelling a uniform inlet, here it is assumed that after some distance the bubbles are uniformly distributed over the cross-section. This will reduce computational times as the flow behaviour close the holes becomes less dynamic.

Figure 14: Photograph of the sparger in the bubble column

3.4.3 Boundary conditions

The inlet gas mass density (g_{mf}, kg/m2/s) is calculated from the inlet gas flow (m3/h) divided by the gas inlet surface area to obtain the gas inlet velocity (v_g, m/s) multiplied with the density of the gas (ρ_g, kg/m3). The number density flux (N, 1/m2/s) is calculated from the gas inlet velocity divided by the volume of a bubble (V_b):

$$g_{mf} = v_g \rho_g,$$

$$N = \frac{v_g}{V_b}.$$

The boundary conditions are given in Table 4.

Table 4: 4 Boundary conditions used in the CFD model

Boundary	Gas	Water	Transport (dissolved hydrogen, bacteria)
Inlet	Gas flux: gas mass density and number density flux	Velocity inlet, depending on batch (0 m/s) or flow through	Concentration of zero
Side walls	No gas flux	No slip	No flux
Top	Gas outlet	Zero pressure	Zero flux

3.4.4 Initial conditions

Flow fields and gas hold-up were set to zero in the initial condition. The pressure was set equal to static pressure distribution over the height. The (dissolved) hydrogen concentration was set to zero and the initial bacteria concentration was set to 0.1 mol/L.

3.4.5 Simulation time

The bacterial growth is a process that occurs over a number of days. Therefore, the simulation time was set to 7 days to try to get a suitable representation of the growth process occurring in the bioreactor. A period of 7 days was chosen as it is in the order of magnitude for the time scale at which maximum growth occurs.

3.4.6 Assumptions

In summary, the most important assumptions in the model are:

- The bubble size remains constant over the height of the system, meaning there is no coalescence or break-up of bubbles.
- Concentration of hydrogen in the bubbles remains constant during the bubble rise. This seems to be a reasonable assumption: in practise, 720 L/h fresh gas is added to the total gas flow of 19000 L/h, so the loss if hydrogen is about 3%.
- The inlet of bubbles is modelled as a uniform inlet over the surface of the sparger. The individual holes in the sparger plate are not modelled in detail (refer to the Proof-of-principle in Section 3.5.1 below).
- For the two-phase system, a bubbly flow interface was used, which means that no separate phase for gas and interface tracking of the bubbles is modelled, but the gas phase is considered as a dispersed phase (see Section 3.4).
- The column is not modelled as a full 3D system, but as a 2D axisymmetric system.

These assumptions were needed to obtain an acceptable computational time for a simulation time of 7 days.

3.5 Results

3.5.1 Proof-of-principle

As a proof-of-principle, a model was developed with a uniform inlet of gas at the bottom and a model for a gas distributor with representative holes size (non-uniform inlet)). The velocity fields, gas volume fractions and dissolved hydrogen concentrations are shown for both models in Figure 15 and Figure 16. The simulations were done for a short simulation time of 60s, so there is no substantial bacterial growth during that short period of time. The velocity fields and gas volume fractions look different using a uniform and non-uniform inlet. The non-uniform inlet shows a more dynamic behaviour, especially at the bottom 50 cm of the reactor. However, the gas volume fractions and dissolved hydrogen concentrations closer to the top become more uniform. A comparison of the models for two gas flow rates using a uniform inlet and a non-uniform inlet at two different gas flow rates is shown in Table 5 and Table 6 respectively (the maximum gas flow rate of 19 m3/h and a lower gas flow rate of 14.4 m3/h is applied for better convergence of the model). Gas hold-up, mass transfer, dissolved hydrogen concentration and bacterial growth (over a short time) are very similar between both systems. As computational times for the uniform inlet system are much smaller compared to the non-uniform inlet (due to the non-uniform hydraulics at the bottom, smaller steps need to be taken by the numerical solver, resulting in much longer calculation times), and results are very similar, the uniform inlet was chosen to model the longer simulation times required for bacterial growth.

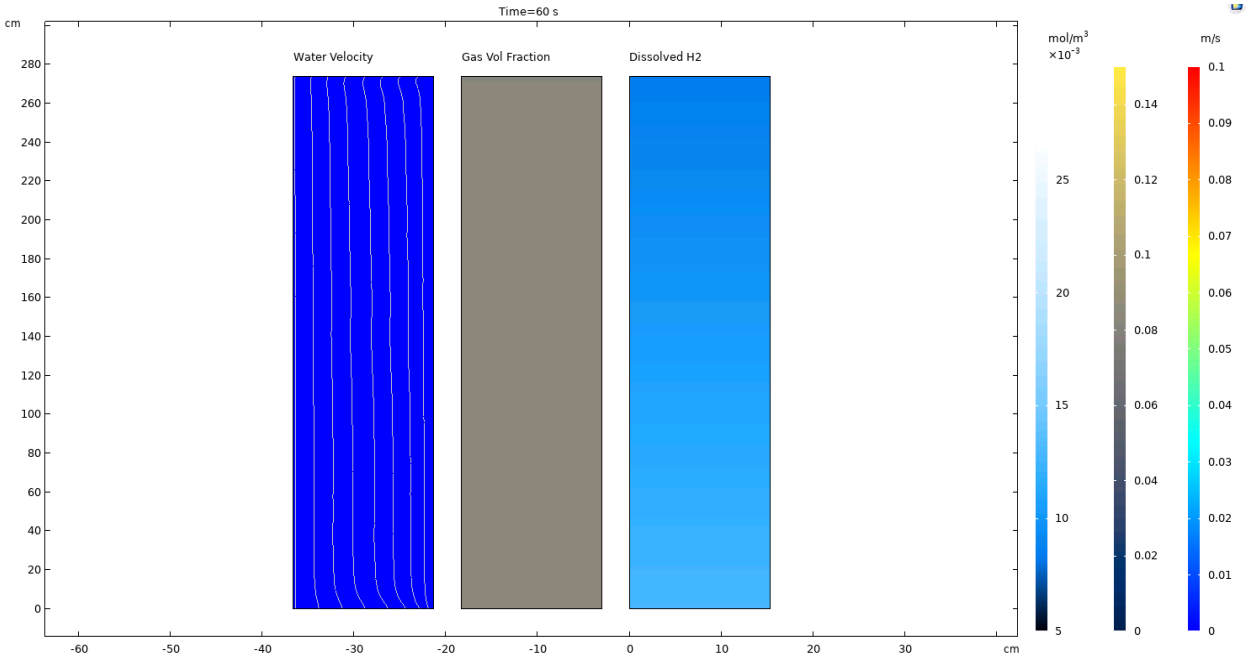

Figure 15: Water velocity, gas volume fraction and dissolved H2 concentrations for a proof-of-principle simulation after 60s using a uniform inlet of gas bubbles (total gas flow of 14.4 m3/h).

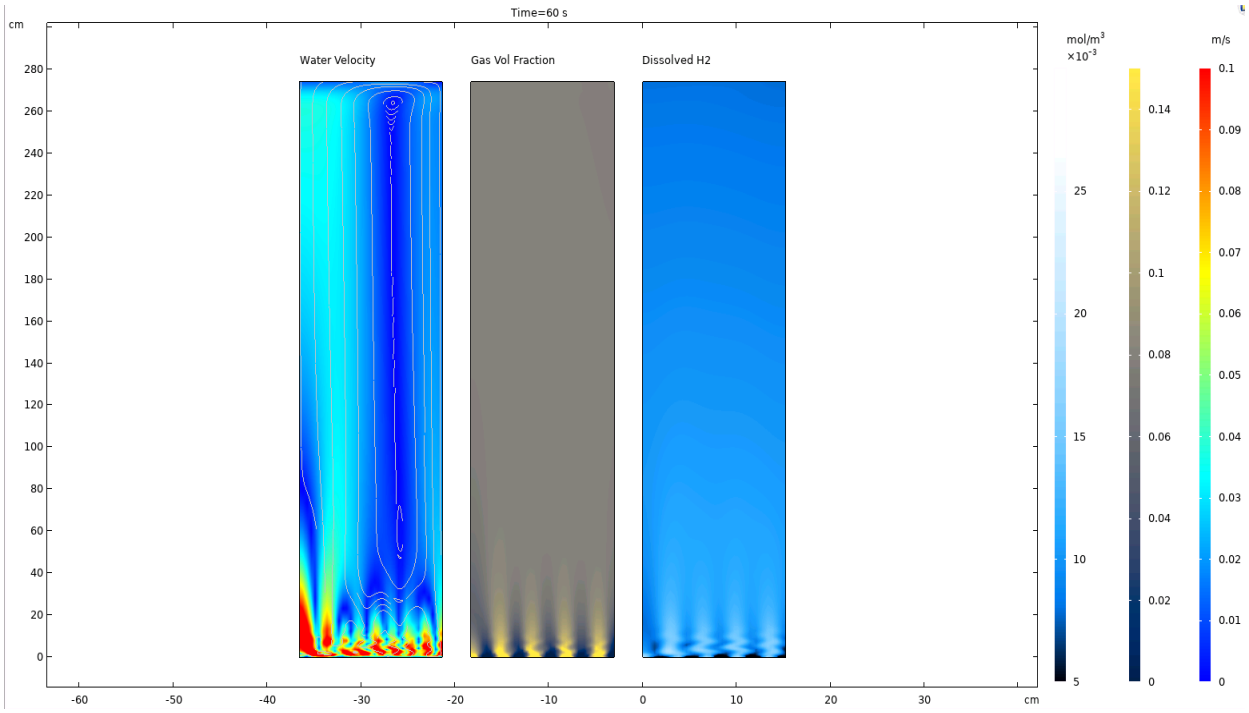

Figure 16: Water velocity, gas volume fraction and dissolved H2 concentrations for a proof-of-principle simulation after 60s using a non-uniform inlet of gas bubbles (total gas flow of 14.4 m3/h).

Table 5: 5 Comparison between uniform and non-uniform inlet at a total gas flow rate of 14.4 m3/h

	Uniform	Holes
Gas hold-up (average)	~8.2%	~7.9%
Mass transfer gas → liquid	8.8^{E-5} kg/s	8.5^{E-5} kg/s
Dissolved H_2 Concentration: at top	8.5^{E-2} mol/m3	8.0^{E-2} mol/m3
Bacterial growth (after 50s) at top	0.253 mol/m3	0.248 mol/m3

Table 6: 6 Comparison between uniform and non-uniform inlet at a total gas flow rate of 19 m3/h

	Uniform	Holes
Gas hold-up (average)	~10.0%	~9.6%
Mass transfer gas → liquid	9.3^{E-5} kg/s	9.0^{E-5} kg/s
Dissolved H_2 Concentration: at top	11.5^{E-2} mol/m3	10.9^{E-2} mol/m3
Bacterial growth (after 50s) at top	0.334 mol/m3	0.332 mol/m3

3.5.2 Calibration and Validation

Only a small amount of measurements were available for calibration and validation of the CFD model as the pilot bioreactor does not have the necessary instrumentation and online sensors to measure some of the required key operating parameters. However, for the bacterial growth model, biokinetic parameters derived from batch and pilot experiments at Avecom were used (see Table 3). The Monod constants were fitted in such a way that the maximum bacterial growth was reached in ~3 days, which was close to the measured values in the pilot reactor.

For validation purposes, the hydrodynamic and biokinetic models could be distinguished. Ideally, the validation of the hydrodynamic modelling is done by measurements of bubble size and dissolved hydrogen concentration. For the time being, only a very straightforward comparison between gas holdup could be made. Both in the model and measurements, the gas holdup was around 8-10% (depending on gas flow rate).

3.5.3 Sensitivity Analysis

A sensitivity analysis was conducted on key operational parameters of interest to further understand the impacts of varying these key parameters on the bacterial growth . This sensitivity analysis was conducted by varying one parameter while the others remain the same as in the reference situation. For the reference situation the following parameter values were used:

- Bubble diameter of 1mm
- Mass transfer coefficient of 6.6E-4 m/s
- Reference pressure of 1.3 bar
- Reactor height and diameter: 2.79m and 0.305m

(a) Mass transfer coefficient

The bacterial growth over the 7 days of simulation is shown in Figure 17. As shown in the graph below with the red line, a higher mass transfer coefficient (k_H = 1E-3 m/s) yields a faster bacterial growth compared with the yield obtained with the reference mass transfer coefficient value of 6.7E-4. But moving from a mass transfer coefficient of 6.7E-4 to 1E-3 the increase in bacterial growth is limited.

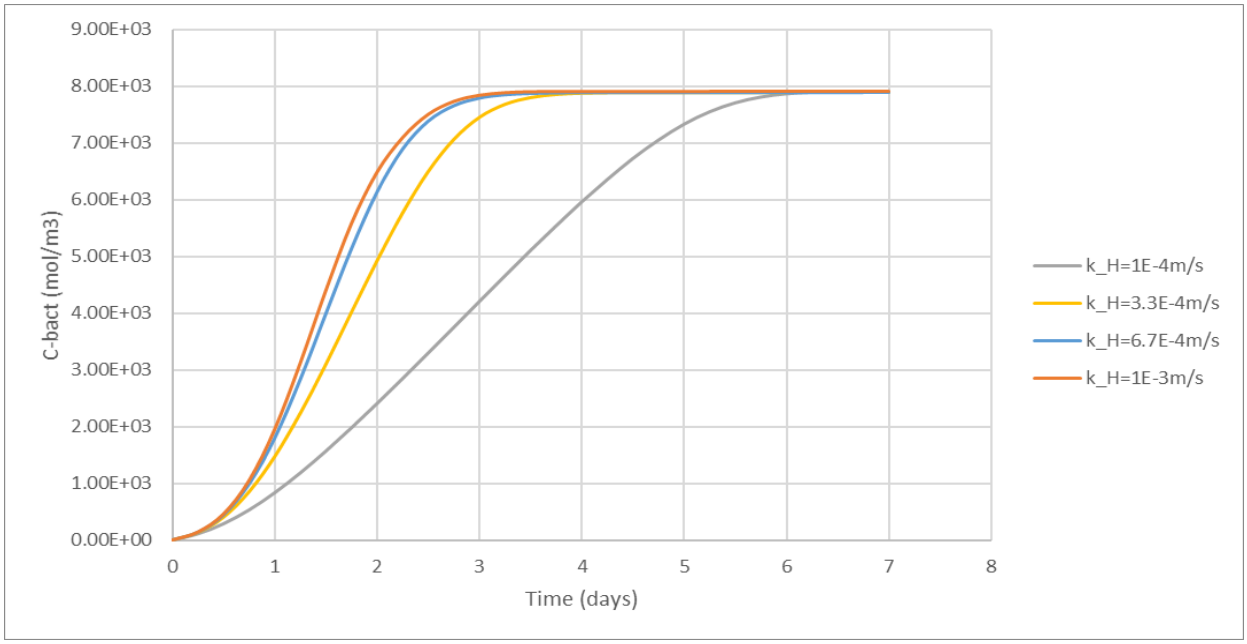

Figure 17: Bacterial growth as a function of time for different mass transfer coefficients.

(b) Bubble size

Small variations in bubble size already have an effect in bacterial growth (see Figure 18). Larger variations in bubble sizes could not be tested, as the numerical model became unstable. Nevertheless, the simulation results suggest that further decreasing the bubble size could further increase the bacterial growth.

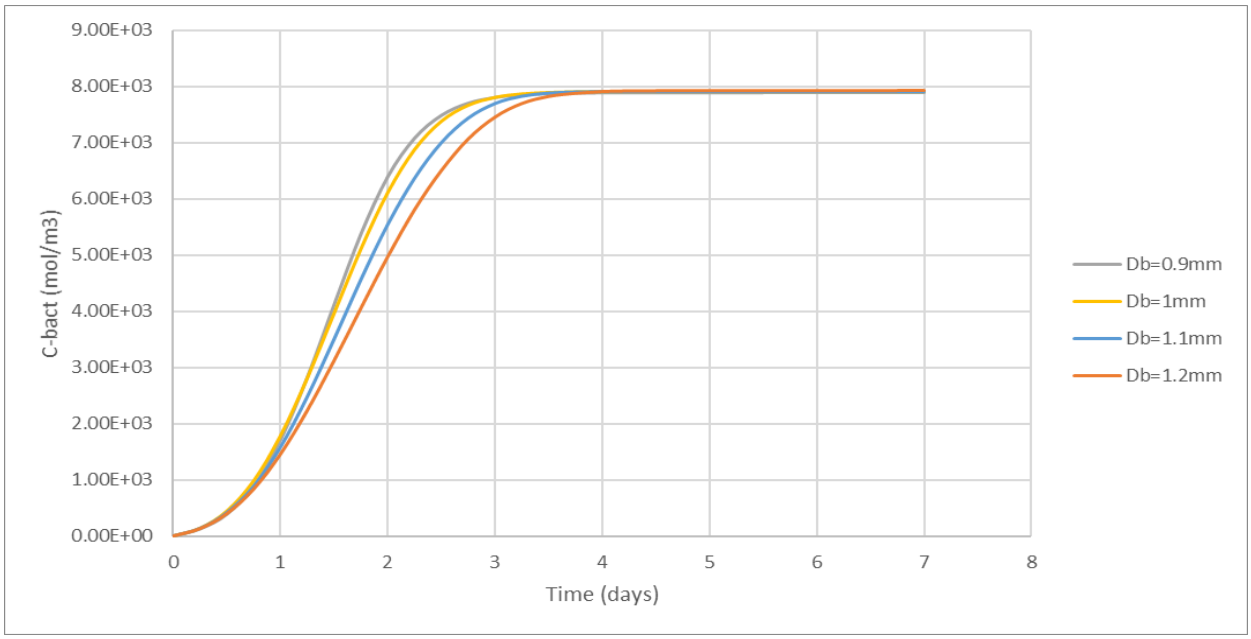

Figure 18: Bacterial growth as a function of time for different bubble sizes.

Also, a first model test was made on an evolving bubble size over the depth. This was implemented by setting the bubble size as a linear function of height. Again, only small changes in bubble size could be modelled. Figure 19 shows a simulation if the bubble size increases from 1mm up to 1.25 mm at the top (yellow line) compared to the situation where the bubble size remained at a constant value of 1mm (grey line). Similar as in the variation of bubble size, the bacterial growth decreases if the bubble size increases over the height of the reactor.

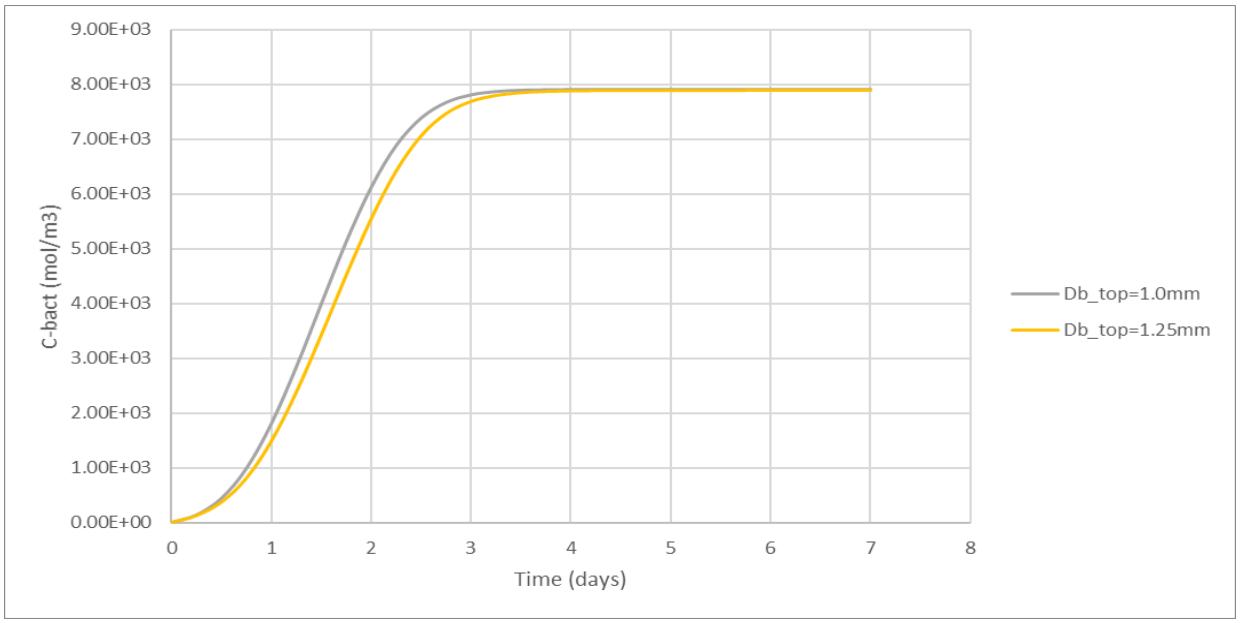

Figure 19: Bacterial growth as a function of time for different bubble size increase over the height of the reactor (Db_top is the bubble size at the top of the reactor)

.

(c) (Over)Pressure

The pressure in the reactor is an important factor for bacterial growth (Figure 2020). A higher pressure means a higher driving force for mass transfer of gas to water (Henry's law), and therefore a higher dissolved hydrogen concentration, which increases bacterial growth. It can be seen that the yield is about 3 times faster when the pressure is increased from 1.3 to 5 bars. However, a remark should be made that the model does not take into account the loss of hydrogen gas in the bubble when the bubble rises. For the reference situation, a calculation was made that this loss is small (about 3%, see Section 3.4.6), however this will be higher at higher pressures.

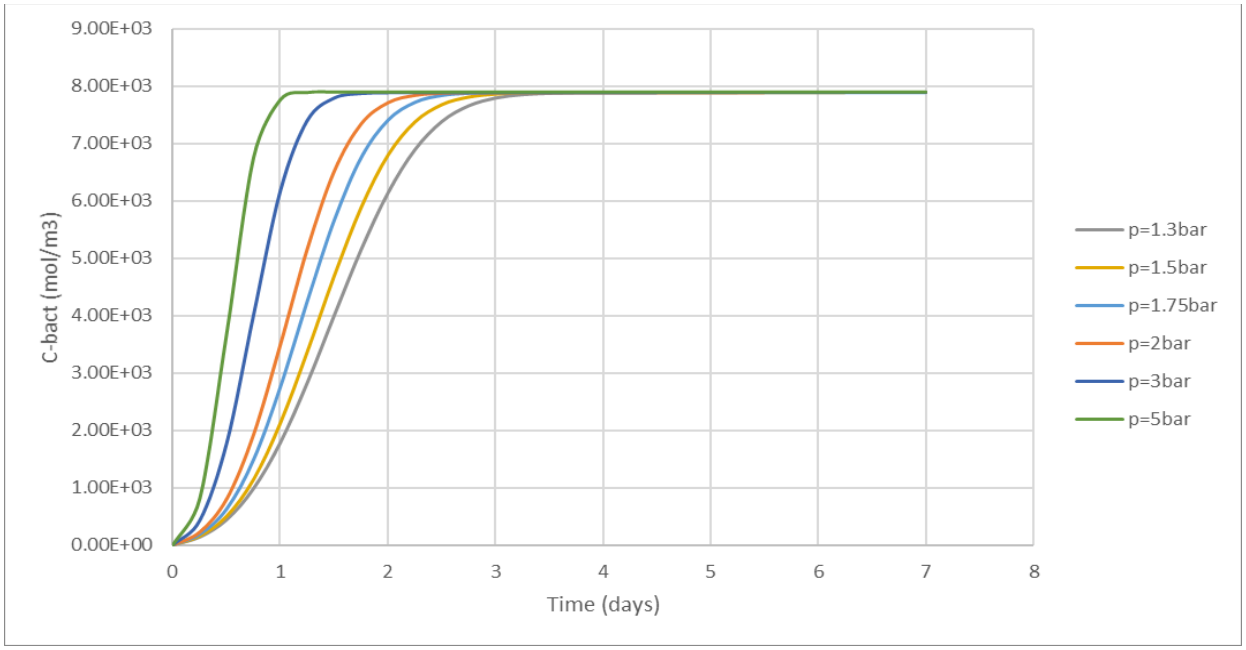

Figure 20: Bacterial growth as a function of time for different pressures of the system.

(d) Height of reactor

The height of the reactor has a small effect on bacterial growth (Figure 21). A higher reactor results in a slightly better bacterial growth, which is mainly caused by increase of hydraulic pressure.

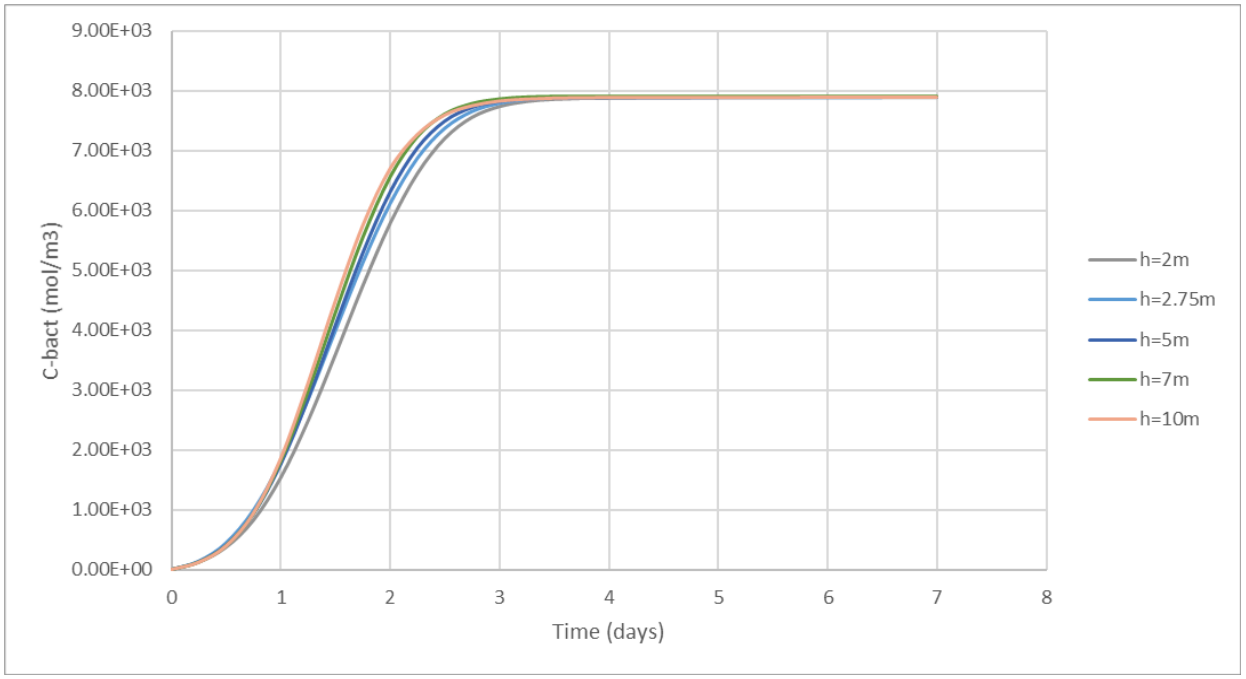

Figure 21: Bacterial growth as a function of time for different heights of the reactor.

(e) Aspect ratio (with same volume)

The aspect ratio (defined as height over radius of the reactor, while keeping the volume at the same value as the reference reactor) is an important factor. A slimmer reactor (higher AR) means a higher bacterial growth (see Figure 22). An optimum is reached at AR of 18 and higher. The AR of the current system is ~18, which is at the region of AR values that increasing AR does not lead to better bacterial growth.

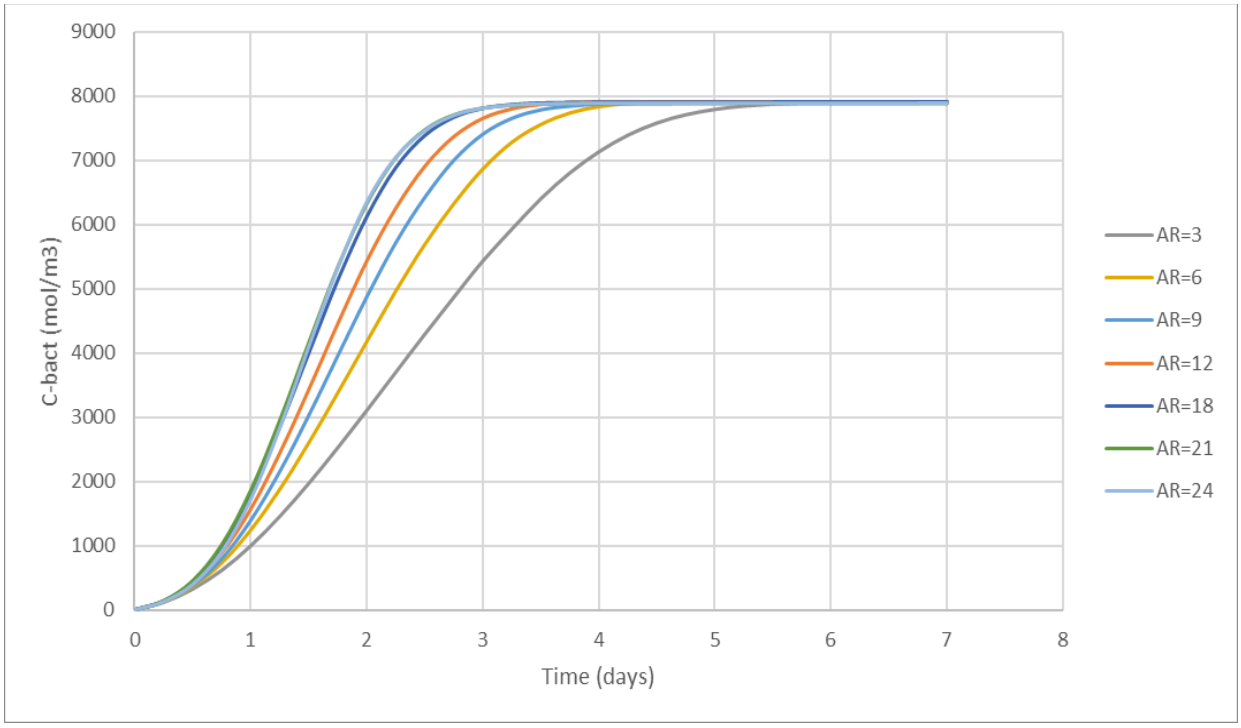

Figure 22: Bacterial growth as a function of time for different aspect ratios of the reactor.

3.6 Fundamental Insights Gained

From the modelling exercise and sensitivity analysis, it was deduced that the most important parameters to improve reactor performance are bubble size, pressure and aspect ratio. According to the model calculations, the aspect ratio is already close to the optimal value at the current reactor design, whereas increasing pressure and decreasing bubble size could lead to a better reactor performance. The modelling results and the sensitivity analysis shone light onto the key parameters that need to be considered and further optimised to improve the performance of the process. The preliminary stage of model development and simulations has offered key insights into the power-to-protein process in a bubble column bioreactor. Obtaining additional operating and monitoring data from the current pilot plant or a scale-up plant will assist in optimising the above key parameters both by improving the bioreactor design and operation as well as further calibrating and validating the CFD model developed.

4 Conclusions and Recommendations

In this study, the aim was to gain fundamental insights into the mass transfer of hydrogen in bioreactors in order to promote the biosynthesis of single-cell protein. More specifically, a literature review together with consultation with experts in the operation of bubble column reactors related to enhancing the mass transfer of hydrogen were conducted. Furthermore, a CFD model using the simulation software COMSOL, was developed and calibrated using the available data to adequately represent the pilot-scale bioreactor that is currently being operated by Avecom to further understand and test the power-to-protein concept.

The following conclusions can be made from the outcome of the above scope of the study.

- The bubble column or slurry bubble column reactors are preferred reactor configurations to achieve a high performance of gas-to-liquid mass transfer. Furthermore, specific to the mass transfer of hydrogen, bubble column reactors are more suitable.

- Consultations with experts in this field also confirm the above conclusion that the bubble column reactor provides the best and right configuration for the power to protein concept.

- These deductions provide additional confidence to Avecom and the project partners on the pilot scale bubble column reactor currently in operation.

- Key design and operating parameters have been identified from the literature and expert consultations for the development of the CFD model to simulate the performance of the bubble column reactor.

- From the modelling exercise and sensitivity analysis, it was concluded that the most important parameters to improve reactor performance are bubble size, pressure and aspect ratio. The aspect ratio is already close to the optimal value (~18) in the model, whereas increasing pressure and decreasing bubble size (<0.9mm) could lead to a better reactor performance. For instance, increasing the pressure from 1.3bar to 5 bars gives a yield 3 times faster.

- The preliminary stage of model development and simulations has offered key insights into the power-to-protein process in a bubble column bioreactor. Obtaining additional operating and monitoring data from the current pilot plant or a scale-up plant will assist in optimising the above key parameters both by improving the bioreactor design and operation as well as further calibrating and validating the CFD model developed.

This study has provided very encouraging outcomes that should provide additional confidence for a scale-up of the power to protein current pilot plant. Based on the results from the model simulations and the conclusions derived, it is recommended to investigate and optimise the key parameters should there be a next phase of the project. For instance, understanding the optimal yield with respect to cost and safety considerations related to increasing reactor pressure and decreasing the bubble size would be important to determine the best economically feasible and viable options to operate the reactor at its optimum protein yield. These key parameters should be considered and closely monitored if a scale-up bubble column reactor is to be considered. Additionally, the current model can be further improved by obtaining the necessary operational data. A suitable network of online sensors to monitor the key parameters and transition to a smarter system would be another next step towards providing sufficient data to further validate, calibrate and fine-tune the model and increase its prediction accuracy.

5 References

- Bouaifi, M., Hebrard, G., Bastoul, D., & Roustan, M. (2001). A comparative study of gas hold-up, bubble size, interfacial area and mass transfer coefficients in stirred gas-liquid reactors and bubble columns. *Chemical Engineering and Processing*, *40*(2), 97–111. https://doi.org/10.1016/S0255-2701(00)00129-X
- COMSOL Multiphysics® v. 5.6. www.comsol.com. COMSOL AB, Stockholm, Sweden.
- Daly, J. G., Patel, S. A., & Bukur, D. B. (1992). Measurement of gas holdups and sauter mean bubble diameters in bubble column reactors by dynamics gas disengagement method. *Chemical Engineering Science*, *47*(13–14), 3647–3654. https://doi.org/10.1016/0009-2509(92)85081-L
- Deckwer, W.D., Cottrell, V., & Field, R.W. (1991). Bubble Column Reactors.
- Deckwer, W. D., & Schumpe, A. (1993). Improved tools for bubble column reactor design and scale-up. *Chemical Engineering Science*, *48*(5), 889–911. https://doi.org/10.1016/0009-2509(93)80328-N
- Degaleesan, S., Dudukovic, M., & Pan, Y. (2001). Experimental study of gas-induced liquid-flow structures in bubble columns. In *AIChE Journal* (Vol. 47, Issue 9, pp. 1913–1931). https://doi.org/10.1002/aic.690470904
- Fan, L. S., Yang, G. Q., Lee, D. J., Tsuchiya, K., & Luo, X. (1999). Some aspects of high-pressure phenomena of bubbles in liquids and liquid-solid suspensions. *Chemical Engineering Science*, *54*(21), 4681–4709. https://doi.org/10.1016/S0009-2509(99)00348-6
- Friedel L, Herbrechtsmeier P, Steiner R. (1980). Mean gas holdup in down flow bubble columns. Ger Chem Eng 3:342-6
- Fujie, K., Takaine, M., & Kubota, H. (1979). *Flow and Oxygen Transfer in Cocurrent*.
- Kantarci, N., Borak, F., & Ulgen, K. O. (2005). Bubble column reactors. *Process Biochemistry*, *40*(7), 2263–2283. https://doi.org/10.1016/J.PROCBIO.2004.10.004
- Hadamard, J. S. (1911). "Mouvement permanent lent d'une sphere liquide et visqueuse dans un liquide visqueux". C. R. Acad. Sci. (in French). 152: 1735–1738.
- Hills, J. H. (1976). The operation of a bubble column at high throughputs: I. Gas holdup measurements. *The Chemical Engineering Journal*, *12*(2), 89–99. https://doi.org/10.1016/0300-9467(76)87002-5
- Hyndman, C. L., Larachi, F., & Guy, C. (1997). Understanding gas-phase hydrodynamics in bubble columns: a convective model based on kinetic theory. *Chemical Engineering Science*, *52*(1), 63–77. https://doi.org/10.1016/S0009-2509(96)00387-9
- Idogawa, K., Ikeda, K., Fukuda, T., & Morooka, S. (1985). Behavior of Bubbles in a Bubble Column Under High Pressure for Air-Water System. *Kagaku Kogaku Ronbunshu*, *11*(3), 253–258. https://doi.org/10.1252/kakoronbunshu.11.253
- Kang, Y., Cho, Y. J., Woo, K. J., Kim, K. I., & Kim, S. D. (2000). Bubble properties and pressure fluctuations in pressurized bubble columns. *Chemical Engineering Science*, *55*(2), 411–419. https://doi.org/10.1016/S0009-2509(99)00336-X
- Krishna, R., De Swart, J. W. A., Ellenberger, J., Martina, G. B., & Maretto, C. (1997). Gas Holdup in Slurry Bubble Columns: Effect of Column Diameter and Slurry Concentrations. *AIChE Journal*, *43*(2), 311–316. https://doi.org/10.1002/aic.690430204
- Kumar, S., Kumar, R. A., Munshi, P., & Khanna, A. (2012). Gas hold-up in three phase co-current bubble columns. *Procedia Engineering*, *42*(August), 782–794. https://doi.org/10.1016/j.proeng.2012.07.470
- KWR 2015.079 (2015), Toepassing van het “Power-to-Protein” concept in de stedelijke watercyclus van Amsterdam, TKI Project

- KWR 2018.078 (2018), Power-to-Protein: eiwitproductie in een circulaire economie, Fase 2 Pilotonderzoek, TKI Project
- Letzel, H. M., Schouten, J. C., Krishna, R., & Van Den Bleek, C. M. (1999). Gas holdup and mass transfer in bubble column reactors operated at elevated pressure. *Chemical Engineering Science*, *54*(13–14), 2237–2246. https://doi.org/10.1016/S0009-2509(98)00418-7
- Li, H., & Prakash, A. (1997). Heat Transfer and Hydrodynamics in a Three-Phase Slurry Bubble Column. *Industrial and Engineering Chemistry Research*, *36*(11), 4688–4694. https://doi.org/10.1021/ie9701635
- Luo, X., Lee, D. J., Lau, R., Yang, G., & Fan, L. S. (1999). Maximum stable bubble size and gas holdup in high-pressure slurry bubble columns. *AIChE Journal*, *45*(4), 665–680. https://doi.org/10.1002/aic.690450402
- Matassaa, S., Verstraete, W., Pikaar, I., Boon, N. (2016) Autotrophic nitrogen assimilation and carbon capture for microbial protein production by a novel enrichment of hydrogen-oxidizing bacteria, Water Research, Vol. 101, pp 137-146.
- Mena, P. C., Ruzicka, M. C., Rocha, F. A., Teixeira, J. A., & Drahoš, J. (2005). Effect of solids on homogeneous–heterogeneous flow regime transition in bubble columns. *Chemical Engineering Science*, *60*(22), 6013–6026. https://doi.org/10.1016/J.CES.2005.04.020
- van Eck, N.J & Waltman, L. (2021). VOSviewer [Computer software]. Centre for Science and Technology Studies, Leiden University, The Netherlands
- Rybczynski, W. (1911). "Über die fortschreitende Bewegung einer flüssigen Kugel in einem zähen Medium". Bull. Acad. Sci. Cracovie, A. (in German): 40–46.
- Sasaki, S., Hayashi, K., & Tomiyama, A. (2015). Effects of liquid height on gas holdup in air-water bubble column. *Experimental Thermal and Fluid Science*, *72*, 67–74. https://doi.org/10.1016/j.expthermflusci.2015.10.027
- Saxena, S. C., Rao, N. S., & Saxena, A. C. (1990). Heat-transfer and gas-holdup studies in a bubble column: Air-water-glass bead system. *Chemical Engineering Communications*, *96*(1), 31–55. https://doi.org/10.1080/00986449008911481
- Schumpe, A., & Grund, G. (1986). The gas disengagement technique for studying gas holdup structure in bubble columns. The Canadian Journal of Chemical Engineering, 64(6), 891–896. https://doi.org/10.1002/cjce.5450640602
- Shah, Y.T., Kelkar, B. G., & Godbole, S. P. (1982). *Design Parameters Estimation for Bubble Column Reactors*.
- Su, X., & Heindel, T. J. (2003). Gas holdup in a fiber suspension. *Canadian Journal of Chemical Engineering*, *81*(3–4), 412–418. https://doi.org/10.1002/cjce.5450810311
- Tsuchiya, K., Furumoto, A., Fan, L. S., & Zhang, J. (1997). Suspension viscosity and bubble rise velocity in liquid-solid fluidized beds. *Chemical Engineering Science*, *52*(18), 3053–3066. https://doi.org/10.1016/S0009-2509(97)00127-9
- *United Nations, Department of Economic and Social Affairs, Population Division (2019). World Population Prospects 2019, custom data acquired via website.*
- Wang, T., Wang, J., & Jin, Y. (2007). Slurry reactors for gas-to-liquid processes: A Review. Industrial & Engineering Chemistry Research, 46(18), 5824–5847. https://doi.org/10.1021/ie070330t
- Xu P. (2020). Analytical solution for a hybrid Logistic-Monod cell growth model in batch and continuous stirred tank reactor culture. Biotechnology and bioengineering, 117(3), 873–878. https://doi.org/10.1002/bit.27230
- Yang, G.Q., Luo, X., Lau, R., Fan, L.S. “Heat-Transfer Characteristics in Slurry Bubble Columns at Elevated Pressures and Temperatures,” Industrial and Engineering Chemistry Research, Vol. 39, (2000), pp. 2568-2577.