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### Accepted Manuscript

Pilot-scale cross-flow microfiltration of *Chlorella minutissima*: a theoretical assessment of the operational parameters on energy consumption

M.L. Gerardo, M.A. Zanain, R.W. Lovitt

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Pilot-scale cross-flow microfiltration of *Chlorella minutissima*: a theoretical assessment of the operational parameters on energy consumption

<sup>a\*</sup>M. L. Gerardo, <sup>a</sup> M. A. Zanain, <sup>a</sup>R. W.Lovitt

<sup>a</sup> Centre for Complex Fluid Processing (CCFP), Systems and Process Engineering Centre (SPEC), College of Engineering, Swansea University, Swansea, SA2 8PP, UK

\*Corresponding author

Email: M.L.Gerardo@Swansea.ac.uk

Michael.L.Gerardo@gmail.com

Tel: +44 1792 60 2303

Fax: +44 1792 295 676

**Abstract** 

Microalgae biomass is seen as a sustainable and socially more responsible feedstock for the

production of biofuels and other fine chemical products. Dewatering algae using membrane

filtration is a leading technology, however the associated costs are typically not determined.

This work investigates the filtration of *Chlorella minutissima* using a pilot-scale cross-flow

microfiltration unit. A filtration model was developed and validated based on permeate flux

as a function of biomass concentration (0.6-19.0 dry cell weight/L) and transmembrane

pressure (ΔP, 1.80-2.10 bar). Processing times for harvesting Chlorella m. were determined

by iteration of the model and costs were related to energy consumption. For the

experimental conditions of 1.95 bar, 1.0 g DCW/L initial biomass concentration, 0.70 kWh,

25 °C and 3.8 m<sup>2</sup> membrane area, harvesting costs were determined as 2.86 kWh/kg

biomass. Subsequent investigation of the influence of the operating parameters and scale-

up effects demonstrated that significant cost reduction to 1.27 kWh/kg biomass was

possible at 1.95 bar, 2.0 g DCW/L initial biomass concentration, 0.46 kWh, 20 °C and 7.6 m<sup>2</sup>

membrane area. Further, biomass concentration was demonstrated to be one of the major

drivers to reduce the cost of harvesting microalgae. Membrane filtration was demonstrated

to be a feasible harvesting process allowing biomass concentrations up to 150 g DCW/L

without using chemicals which complicate the downstream processing stages.

**Keywords:** harvesting, microalgae, pilot-scale, microfiltration, energy

2

#### 1. Introduction

Through a photosynthetic process, microalgae are able to uptake metals and nutrients such as carbon, nitrogen and phosphorous for their growth and transform these materials into valuable commodities such as proteins, sugars, lipids and silica. Whilst the algal proteins are the drive for microalgae as feed and "foodstuff", the biofuel focus is, on the other hand, related to the level of lipids which may be chemically transformed into biodiesel [1]. Given that arable land is not required for the production of such biomass, microalgae is considered to be a more responsible feedstock for the production of biofuels [2]. Indeed, efforts have been made to realise this potential and thus researchers have reported on the feasibility and limitations of integrating microalgae cultivation with wastewater treatment [3-5] and CO<sub>2</sub> mitigation [6-8]. Nonetheless, current costs of producing algal-oil are still prohibitive at \$2.80/L [9], and thus researches are also focusing on using microalgae in the food, feed, chemical and pharmaceutical sector[10]. Nonetheless, more recent estimates have demonstrated that there is an added cost benefit on reduction of emissions and resource availability [11].

Microalgae are unicellular organism with cell sizes ranging from 3-30  $\mu$ m and typically grow at biomass concentrations below 1.0 g dry cell weight per litre (DCW/L). One of the drawbacks of microalgae technology is the dewatering process which reduces the very dilute culture to more convenient concentrations, essential for low cost operations. Most common separation technologies associated to microalgae harvesting are sedimentation, centrifugation, coagulation-flocculation, dissolved air flotation and filtration. Taking advantage of the relatively low energy input and chemical-free separation, membrane filtration seems to be one of the leading technologies for harvesting microalgae. Generally, given the correct membrane is selected, membrane filtration allows nearly complete cell

recovery and up to 15 % solids whilst process media can also be recovered for reuse. For that purpose several membrane configurations exist, namely spiral-wound, plate and frame, tubular, and fiber [12, 13]. The exact influence on the harvesting performance and associated costs is yet to be established. At pilot and industrial-scale, membrane filtration systems are most commonly operated in a cross-flow mode which minimises the build-up of cake. Nevertheless, most work on microalgae harvesting has been carried at very small scale and typically do not determine energy inputs during the harvesting process. Such energy requirement for microalgae harvesting is vital in assessing the feasibility of membrane filtration in such applications. Mohn (1980) reported on the energy requirement for five different vacuum filter units to be 0.1-5.9 kWh/m<sup>3</sup>. However, there is currently focus on different algal species and thus these technologies do not represent the current processing requirements [14]. More recently, theoretical estimations of energy input have been attempted for lab-scale processes. Power consumptions for microfiltration systems were estimated at 0.169 kWh per kg microalgae and 0.3-0.7 kWh/m<sup>3</sup> [15, 16]. Disappointingly, no indication of the energy determination methodology was given. Only recently, energy requirement for harvesting microalgae using microfiltration has been reported. Upon defining the filtration properties at pilot-scale, the authors determined harvesting costs of Scenedesmus species to be 2.23 kWh/m<sup>3</sup>. A systematic iteration of the filtration model allowed detecting potential cost reduction to 0.90 kWh/m<sup>3</sup>[17]. Nevertheless, this work was limited to one species and may misrepresent the costs associated with harvesting other microalgae species. Energy requirements are of general importance to filtration processes, as pumping costs contribute significantly to overall operating costs. Energy requirements are especially important for algal biofuels where energy sustainability aspects must be addressed. Here, very low-energy is critical for first stage harvesting technologies. Energy is

less critical for second or thirdstage harvesting, where feed streams contain higher concentrations of algae and the objective is more focused on low-cost technologies that can achieve final concentrations in the range of 15% DCW. Energy requirements are less critical for high-end products such as pharmaceuticals, where profits can more easily absorb energy costs. Energy requirements help to identify products that can most benefit from filtration also leading to overall energy savings by replacing other technologies in the production process [17, 18].

Chlorella minutissima is a seawater species and has advantage over other species including fast growth and ease of cultivation. Moreover, this species has a high level of amino acids and polyunsaturated fatty acids, which are potentially important materials in health foods and pharmaceuticals [19-21].

In this work we have investigated the harvesting of *Chlorella minutissima* using a pilot-scale cross-flow microfiltration unit. A filtration model was developed which enabled a genuine prediction of the harvesting times as a function of the operational parameters such as transmembrane pressure ( $\Delta P$ ), initial biomass concentration, membrane area and temperature. Energy consumption was directly measured and related to processing times, harvesting costs were determined and compared to that of *Scenedesmus species*.

#### 2. Materials and methods

#### 2.1. Cultivation and characterisation of Chlorella minutissima

Chlorella m. was cultivated as a monoculture in a 600 L closed-loop photobioreactor (Biofence<sup>TM</sup>, Varicon Aqua Solutions, UK) until stationary stage using commercial f/2 medium as defined in table 1 (Varicon Aqua Solutions, UK). The microalgae were harvested after 18 days directly from the photobioreactor at a concentration of 0.52 g DCW/L. The

microalgae particle size distribution was determined by dynamic light scattering (Malvern Mastersizer 2000MU) and surface charge was determined bymeasurement of electrophoretic mobility (Malvern Zetasizer2000). *Chlorella m.* cells were imaged using scanning electron microscope(Hitachi S4800, Swansea University). A second batch ofmicroalgaewas used to validate the filtration model. In a 2000 L photobioreactor (built insitu), *Chlorella m.* was also cultivated in f/2 nutrient media until stationary phase of which 800 L were used in the validation experiment.

#### 2.2. Pilot-scale cross-flow microfiltration rig

The pilot-scale filtration rig consisted of a 200 L stainless steel tank connected to a centrifugal pump (LowaraSV408) as shown on Figure 1. A spiral wound membrane (Koch part number 3838-K618-HYT) with 3.8 m<sup>2</sup> and 0.1 µm pore size was fitted in a 4in. diameter stainless-steel shell. A series of 1.5in.stainless steel pipes connected the parts in a closed loop so that the retentate returnedback to the feed tank. The system also featured two diaphragm pressure sensors rangingfrom 0 to 6 bar, two temperature sensors ranging from 0 to 60°C, a flow meter and a permeate flow meter Nixon-type 100/200 ranging from 100 to 10000 L/h. A stainless steel coil was submerged in the feed tank which provided temperature control at 20 °C. A bypass valve was used to maintain the cross-flow velocity constant at 1.01 m/s (flow was 4.24 m<sup>3</sup>/h through a cross sectional area of 11.4 cm<sup>2</sup>) since this velocity was feasible at the different pressures at pump flow was a fixed rate. The pilotscale system was supported by a 2x4 in. stainless steel frame and was mounted on castors. Back flushing capability was not present. Routine cleaning of the membrane was typically performed by rinsing with tap water and chemical cleaning. For that purpose, two to three tank volumes of tap water were used to flush the system and thereafter a chemical cycle of

30-60 minutes at pH 11 using NaOH in a closed system (permeate returned back into the feed tank) were carried out as routine procedure.

#### 2.3. Filtration studies

The investigation of the filtration characteristics of *Chlorella m.* was performed in relation to biomass concentration across the range 0.52-4.02 g DCW/L under constant velocity and temperature. At different biomass concentrations, permeate flux was monitored as a function of pressure ( $\Delta P$ ) across the range 1.80-2.10 bar as per membrane specification on operating parameters (Koch KMS MFK Food & Dairy MF elements). All filtration experiments were operated within the pressure control region. Biomass was measured spectrophotometrically (UNICAM UV 300) at 750 nm against a DCW calibration curve. Darcy's general membrane equation was used to establish the dependence of flux (J) as a function of cake resistance ( $R_c$ ):

$$J = \frac{|\Delta P| - |\Delta \Pi|}{(R_m + R_c + R_c)_{\text{II}}}$$
 (Equation 1)

 $J - permeate flux in L/(m^2.h)$ .

 $\Delta P$  – transmembrane pressure in bar.

 $\Delta\Pi$  – osmotic pressure in bar.

R<sub>m</sub> – resistance to flux owing to intrinsic membrane resistance in m<sup>-1</sup>.

R<sub>c</sub> – resistance to flux owing to cake deposition in m<sup>-1</sup>.

 $R_f$  – resistance to flux owing to gel foulants deposited and adsorbed on the membrane surface in  $m^{-1}$ .

 $\mu$  – viscosity of the permeate fluid in Pa.s.

In the case of microfiltration systems, both  $\Delta\Pi$  and  $R_f$  are considered negligible. Intrinsic membrane resistance ( $R_m$ ) was calculated in relation to clean water flux,where  $R_c$  is zero, across the range 1.80-2.10 bar.

Once the filtration model was developed, validation of the model was performed with a second batch of microalgae, in which a total of four tank volumes (~800 L) were used, batch 1, 2, 3 and 4, respectively.

#### 2.4. Energy consumption and associated costs

Energy consumption was directly measured in kilowatt hours using a handheld electricity meter Efergy model Elite 1.0T (Efergy, Sheffield, UK). When in operation, the pilot-scale filtration system was using 0.70 kWh. According to the manufacturer, the maximum pump efficiency for the LowaraSV408 1.5 kW centrifugal pump is 0.58. For such hydrodynamic conditions (4.24 m³/h at 3.5 bar) industrial pumps can be up to 0.85 efficient and thus scale up effect could reduce energy intake to 0.46 kWh.

Upon modelling the filtration process, to facilitate the determination of the operating parameters on the harvesting costs, processing times were determined for each case scenario: (1) experimental conditions (2) increase of  $\Delta P$ , (3) increase of initial concentration, (4) increase in membrane area and (5) increase in temperature.

The time necessary to process the total volume is given by

$$\frac{dV}{dt} = J \times A \tag{Equation 2}$$

dV – volume differential at a certain moment.

*dt*– time.

J –permeate flux.

A – membrane area.

Integrating under steady-state conditions, i.e.  $\Delta P$  and  $\mu$  are constant, equation 2 becomes

$$V_t = V_{inital} - J_t \times A \times t \tag{Equation 3}$$

 $V_{initial}$  – total initial volume being processed.

 $V_t$  – volume remaining at instant t.

 $J_t$ —instantaneous permeate flow rate at timet.

The viscosity of water-like fluids is a function of temperature given by the Lewis and Squires method[23]

$$\mu = 0.00002414 \times 10^{\left(\frac{247.8}{T-140}\right)}$$
 (Equation 4)

T – temperature in K.

Processing times were determined by iteration steps using equation 3 at every 10 second time intervals until a remaining volume of 10 L. A detailed procedure can be found elsewhere [17]. This method accounts for the continuous loss of flux as the biomass concentration increases with time. Finally, energy consumption and processing costs were compared to that previously published for *Scenedesmus species*.

#### 3. Results and discussion

#### 3.1. Characteristics of Chlorella minutissima

Chlorella m. is a unicellular spherical cell as shown in Figure 2A. This type of seawater organism does not form colonies and thus particle size distribution is relatively narrow. Cell size distribution was determined as 2.28-4.44  $\mu$ m with an average cell diameter of 3.11  $\mu$ m(Figure 2B). Particle size was also confirmed by SEM where very similar cell diameters

were observed. Surface charge was determined in relation to electrophoretic mobility. At pH 8.5, surface charge was determined to be -9.1 ( $\pm$ 1.6) mV and pH did not seem to greatly influence surface charge. At pH 4 and pH 10, surface charge was -7.7 ( $\pm$ 1.5) and -7.6 ( $\pm$ 2.0) mV, respectively. Such low variation of the surface charge with pH is most likely due to the presence of extracellular organic matter (EOM) present on the cell's surfaces which act as a buffer.

#### 3.2. Pilot-scale filtration studies

The investigation of the filtration properties of *Chlorella m.* was set upon measuring permeate flux as a function of both biomass concentration and ΔP. Nevertheless, filtration is a dynamic process owing to the continuous build-up of cake resistance. Constant hydrodynamic conditions may be achieved when permeate flux is constant. Illustrated in Figure 3 is the decline in permeate flux at fixed concentration of 0.52 g DCW/L. During the first few minutes of filtration the flux drops drastically from 102.6 LMH to ~80 LMH, until reaching steady-state after around 120 minutes. Steady-state flux was measuredas 65.8 LMH which represented a loss in flux of 36 % at constant concentration and hydrodynamic conditions. Such drop in flux was a result of the development of a fouling layer on the membrane surface. Fouling phenomena during filtration of microalgae takes place by a conjunction of factors: absorption of EOM onto the membrane surface, pore clogging by the microalgae cells and consequent deposition of a cake layer [24, 25].

The filtration model developed in this work for *Chlorella m.* was based on the biomass concentration as dry cell weight (DCW)since a detailed investigation of the fouling phenomena was not carried out. Upon achieving the constant hydrodynamic conditions at

constant concentration (Figure 3), permeate flux was measured across the pressure control region of 1.8-2.1 bar at different biomass concentrations (Figure 4).

Clean water flux varied between 76.0-97.4 LMH across the  $\Delta P$  range 1.80-2.10 bar. In general, across the biomass concentration range investigated,  $\Delta P$  had a greater influence on the permeate flux at lower biomass concentrations. At biomass concentrations above 3.14 g DCW/L,  $\Delta P$  did not exert such influence since flux was less dependent on pressure. Nevertheless, permeate flux was highly dependent on biomass concentrationas higher flux was possible at lower concentration. Indeed, across the  $\Delta P$  range of 1.8-2.1 bar,  $R_c$  increased linearly as a function of biomass concentration until 3.14 g DCW/L (Table 2). Between 0.52 g DCW/L and 3.14 g DCW/L,  $R_c$  increased sharply until becoming concentration independent at around 3.14-4.03 g DCW/L. In this case, the rate of deposition of cells onto the cake layer was equal to that being removed by the crossflow suspension, thus leading to little variation of  $R_c$  at higher biomass concentrations. As a result, higher biomass concentrations positively affected the removal of the cake layer which is similar to that previously reported elsewhere [26]. Figure 4 illustrates the influence of  $\Delta P$  on permeate flux at different biomass concentrations.

#### 3.3. Modelling and validation of the filtration

The modelling of the microalgae harvesting process reflects the permeate flux profile in relation to biomass concentration. Using the slopes from the linear regressions  $R_m$  and  $R_c$  were calculated from Equation 1. Table 2 summarises the data where the logarithmic regression was the best fit (Equation 5, Table 2) and was the basis for the model.

Once the model was established, theoretical flux was determined by successive iterations using Equation 5 (see Table 2) and Equation 1 at a given pressure and temperature as a

function of microalgae biomass concentration. The validation of the model consisted of the measurement of permeate flux and biomass concentration during harvesting of 800 L of *Chlorella m.*, which equated to four tank volumes. Figure 5 illustrates both these settings with good agreement up to 19.0 g DCW/L. Although the model was originally developed as a function of biomass concentration up to 4.0 g DCW/L, flux prediction was possible and in agreement to that experimentally observed up to 19.0 g DCW/L. In fact, at concentrations of 5.0 g DCW/L and above, batches 3 and 4 seem to move towards the same fluxes predicted by the theoretical model.

#### 3.4. Energy consumption and associated costs

Based on the *Chlorella m.* harvesting model, flux prediction was possible under a variety of operational conditions by simultaneously iterating equations 1, 3, 4 and 5. Once the time needed for the harvesting of a certain volume of microalgae is known then energy consumption is determined based on the total operation time and energy intake. At the experimental conditions (Figure 5), overall cost of harvesting *Chlorella m.* was 2.86 kWh/kg biomass obtained. The increase of  $\Delta P$  from 1.95 bar to 2.10 bar would allow a potential saving in processing time with a harvesting cost of 2.72 kWh/kg biomass. Even though it is possible to operate at higher  $\Delta Ps$ , the fouling phenomenon would be enhanced due to the higher rate of particle deposition on the membrane surface. In cross-flow filtration, cake formation is minimised by the tangential velocity which directly relates to the fluid velocity generated by the pump. Therefore, the increase in  $\Delta P$  leads to an increase of the pump requirement and resulting costs. Moreover, higher tangential flow generates higher shear flow which enhances the transmission of intracellular matter and increased fouling of the membrane [25]. As illustrated in Table 3, the increase of the initial biomass concentration

and membrane area greatly influenced the harvesting costs. Even though at an initial biomass concentration of 2.0 g DCW/L processing time is higher, the quantity of biomass present at the end of harvesting is superior. Thus a reduction of the operational costs from 2.86 kWh to 1.93 kWh/kg biomass would be attainable, however capital costs would increase. Doubling the membrane area from 3.8 m² to 7.6 m² would most certainly reduce operational costs associated with energy consumption. In this case scenario a reduction of harvesting cost to 1.43 kWh/kg biomass could be possible. Nevertheless, such case scenario assumes that the same hydrodynamic conditions are identical to that for 3.8 m² membrane area and this approach is only valid from a theoretical modelling standing point. Finally, an increase of the process temperature from 20 °C to 30 °C would lead to a potential cost reduction from 2.86 kWh to 2.27 kWh/kg owing to reduced viscosity of the microalgae suspension (Equation 4). Nevertheless, lower temperatures are likely to be preferred for the preservation of the microalgae components. The analysis of the culture temperature is likely to be of interest when comparing regions of lower and higher latitudes where significant temperature gradients are expected to take place over the year.

Optimal operating conditions have also been considered. These account for scale up effects, improved process efficiency given by higher membrane area, reduced biomass cost from greater initial biomass concentration and biomass preservation given at 15 °C. Under such operating parameters *Chlorella m.* harvesting costs were determined as 1.27 kWh/kg biomass obtained.

As highlighted by the filtration model, operating parameters have a great influence on the processing costs associated with energy consumption. Membrane area and initial biomass concentration were demonstrated to have a great impact on such costs. Wherever

applicable and financially feasible, increasing the membrane surface area and improved microalgae cultivation strategies could be the best strategy for optimal harvesting. In contrast with harvesting *Scenedesmus sp.* by microfiltration, *Chlorella m.*required more energy for the harvesting step. Under any case scenarioin Table 3, the freshwater microalgae *Scenedesmus sp.* had a lower energy requirement. This is a result of the larger particle size of *Scenedesmus sp.*, 2.94–10.60 μm with a mean particle diameter of 5.58 μm. Such larger particle size is likely to result in less cake resistance which facilitates the filtration process. In addition, surface colloidal chemistry also plays a fundamental role in such phenomena. The negatively charged cell wall surface of microalgae cells repel one another, resulting in increased cake porosity as a function of the repulsive interaction energy. Indeed, surface charge of *Scenedesmus sp.* was reported to be -25.2 mV at pH 8.5[17]against -9.1 mV for *Chlorella m.* at pH 8.5.

3.5. Feasibility of membrane filtration and downstream processing requirements

Cross-flow microfiltration proved to be a feasible technology to harvest *Chlorella m.* given that permeate fraction was absent of microalgae cells (optical density was below 0.01 at 750 nm) and thus maximizing cell recovery. Such separation process may be operated up to a maximum of 15 % solids (~150 g DCW/L, typical maximum solids concentration for feasible membrane filtration) and thus there is scope for further concentration of microalgae biomass than that here reported. For a mass balance of 2000 L of microalgae suspension, it is possible to determine processing costs in relation to different initial biomass concentrations by iteration of the harvesting model developed. In order to avoid cavitation of the pump, the minimum holding volume of the filtration system was defined as 10 L and thus volumetric concentration factors (VCF) are a function of both maximum biomass

concentration and minimum holding volume possible. The determination of energy consumption takes in account optimal operating parameters: ΔP of 1.95 bar, temperature of 20 °C, membrane area of 7.6 m<sup>2</sup> and pump efficiency of 85 %. Table 4 highlights that across the initial biomass range 0.5-2.0 g DCW/L, VCF differ significantly affecting also biomass harvesting costs. While such filtration system may be operated up to 150 g DCW/L, the VCF permissible is very much dependent upon the initial biomass concentration. Indeed, at concentrations of up to 0.75 g DCW/L a VCF of 200 is possible. On the other hand, when harvesting 2000 L of Chlorella m. at 1.0 and 2.0 g DCW/L, VCF would be 133.3 and 75.0, respectively. Seemingly, lower biomass concentrations lead to higher VCF and lower energy consumption owing to lower biomass content in the microalgae suspension. Conversely, higher initial biomass concentration limits the VCF and lead to higher processing times due to increased biomass load. Nevertheless, final harvesting costs follow an inverse trend since at the end of each batch different amounts of biomass would be present. Although, in theory energy consumption may be minimised, higher initial biomass concentrations are required in order to reduce final product costs which has also been noted by other authors [18].

Very few cost estimated can be found in the literature and these are typically associated to laboratory scale systems. Even so, the available cost estimates in the literature for harvesting microalgae by membrane filtration are summarised in Table 5.

For comparison purposes, the energy consumption for harvesting *Chlorella m.* was extracted from Table 3 considering an initial concentration of 0.75 g DCW/L which was a similar concentration to that used in the other studies. It is noted that energy consumption for harvesting *Chlorella m.* (1.3 kWh/m³) was similar to that for *Scenedesmus sp.* (0.9 kWh/m³), however much lower energy consumption was reported for both forward osmosis (FO, 0.3

kWh/m<sup>3</sup>) and magnetically induce membrane vibration (MMV, 0.2 kWh/m<sup>3</sup>). Such lower energy consumption was related to the very low operating range of biomass concentration. Moreover, the harvesting costs of Chlorella m. was slightly higher than that reported by Bhaveet al. (2012) for Nannochloropsis yet lower than that reported by Danquah et al. (2009). As demonstrated by this work, harvesting costs may depend to a great extent on the initial and final concentration of the microalgae suspension and process volume. In addition, the variety of membranes which may be operated at different operating settings associated with the diversity of microalgae species, highlights that each microalgae species may represent a unique set of challenges with specific harvesting costs. Harvesting microalgae up to 50-150 g DCW/L using membrane technology is ideal and has relatively low energy consumption. In contrast, centrifugation is potentially more applicable to achieve final biomass concentrations up to 100-250 g DCW/L, however this option carries a high energy consumption. It is likely that in some case scenarios, a synergetic approach between microfiltration and centrifugation will represent the best harvesting strategy for microalgae. Most importantly, the selection of the harvesting technology is undeniably linked to the downstream processing (DSP) requirements. Microalgae drying processes such as freeze-drying need a minimum amount of water to be more effective, on the other hand spray-drying allow an ideal feed content of 5-10 % solids. Microalgae cell disruption is also carried at relatively low biomass concentrations (5-20 % solids) where it is expected that product recovery is easier at lower biomass concentrations (<15 % solids) [30]. Even though there is no agreement in the literature, a variety of lipid extraction technologies reported do not refer to highly concentrated microalgae slurries and work at rather low concentrations such as 0.5-10 % solids [31-33].

Membrane filtration is set as one of the leading technologies for the harvest of microalgae. The separation mechanism is solely based on size and benefits from no addition of chemicals which potentially complicate the DSP of microalgae products or reuse of the process water. Although the efficiency and final costs may be species dependent, the feasibility of harvesting microalgae using membrane technology is expected to be applicable across the entire range of species regardless of the biochemical properties. The work developed herein demonstrates the energy consumption associated to microalgae harvesting using membrane filtration. On an energy basis, microalgae calorific values range from 18-28 MJ/kg biomass depending on the biochemical composition [34, 35]. Given the calculations on energy consumption provided herein, harvesting of microalgae has an associated energy 10.29-3.51 MJ/kg depending on the chosen conditions thus harvesting of microalgae may represent 57.2-12.5 % of the embedded energy in the microalgae biomass. Other associated costs such as membrane, cleaning and lifetime are also important and were not considered here. Additionally, no hardware for back-flushing of the membrane was available which is commonly used to maintain higher operating fluxes potentially leading to higher performance. Finally, a holistic analysis, such as a life cycle analysis, on the use of membrane technology for harvesting microalgae is currently missing

#### 4. Conclusions

in the available scientific literature.

The filtration characteristics of *Chlorella minutissima* were investigated using a pilot-scale microfiltration system. A model was developed and showed good correlation with experimental data and thus a theoretical analysis of theenergy consumption were determined for different case scenarios. The optimal case scenario which considered scale-

up effects, improved microalgae cultivation strategies for higher biomass concentration and increased membrane area led to a minimal harvest cost of 1.27 kWh/kg microalgae. A theoretical modelling approach has identified opportunities for 57 % reduction of the experimental harvesting costs, nonetheless these calculations does not dispense experimental validation.

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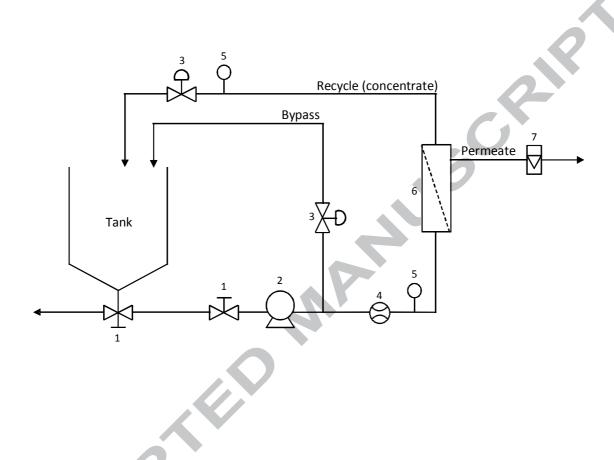
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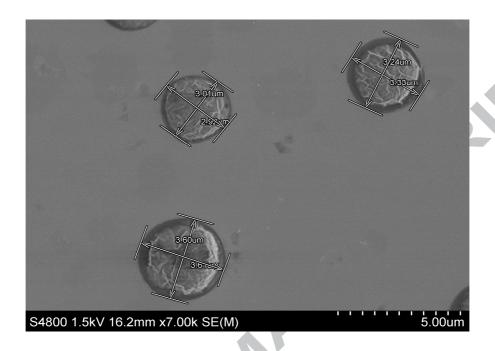
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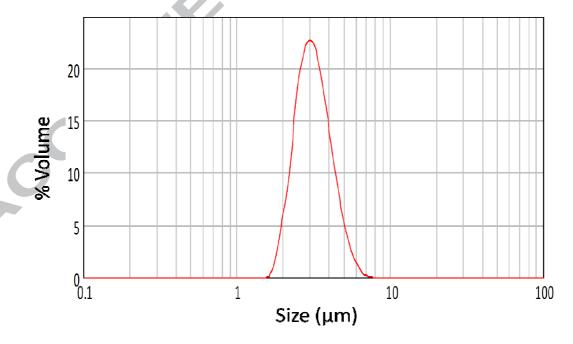
**Figure 1.** Schematic diagram of the microfiltration membrane system. 1 – diaphragm valve, 2 – centrifugal pump, 3 – butterfly valve, 4 – flow meter, 5 – pressure and temperature probes, 6 – membrane filter, 7 – permeate flow meter.



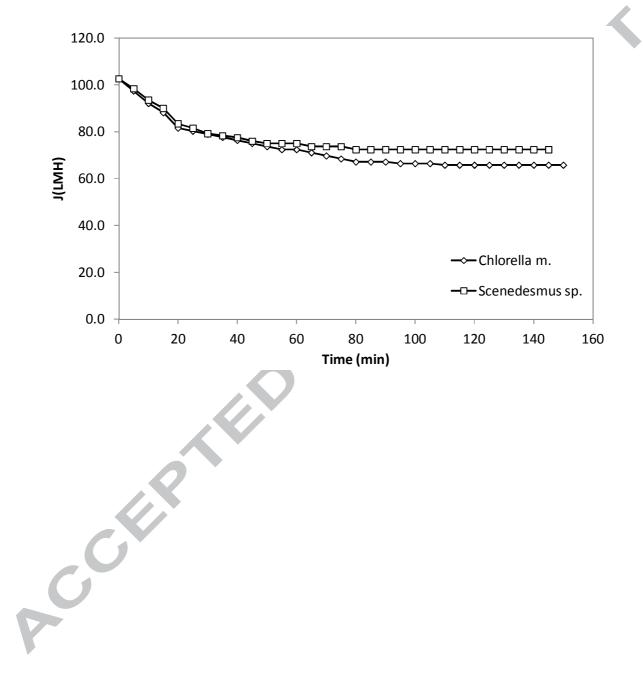
**Figure 2A.** SEM image of *Chlorella minutissima* cells dried at room temperature for at least 48h in a desiccator.



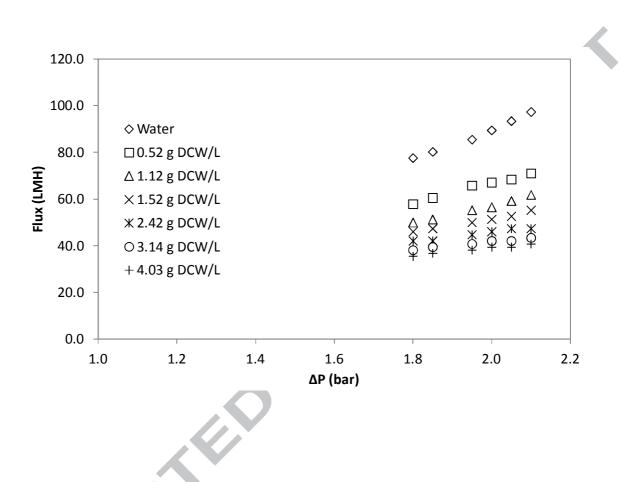
**Figure 2B.** Particle size distribution of *Chlorella minutissima* at stationary stage measured as particle volume distribution.



**Figure 3.** Decline in permeate flux with time under constant concentration of 0.52 g DCW/L (*Chlorella m.*) and 0.85 g DCW/L (*Scenedesmus sp.*) at  $\Delta P$  1.95 bar, 20 °C and 1.01 m/s.



**Figure 4.** Influence of the biomass concentration on cake resistance ( $R_c$ ) obtained by pressure excursions at 1.80< $\Delta$ P<2.10 bar, 20 °C and 1.01 m/s under steady-state conditions.



**Figure 5.**Model validation using experimental data from four consecutive tank volumes of *Chlorella minutissima*. Experimental conditions: 0.64 g DCW/L (initial concentration), 20  $^{\circ}$ C,  $\Delta P$  was 1.95 bar and  $R_m$  2.54E+12  $m^{-1}$ . Cross-flow velocity was maintained constant at 1.01 m/s. Biomass concentration was monitored spectrophotometrically at 750 nm.

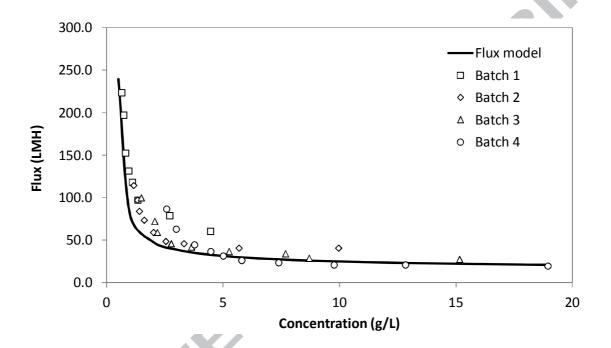


Table 1. Composition and concentration of the f/2 growth medium according to [22].

Component	Concentration (mM)				
NaNO <sub>3</sub>	8.20x10 <sup>-1</sup>				
NaH <sub>2</sub> PO <sub>4</sub> .H <sub>2</sub> O	3.62x10 <sup>-2</sup>				
Na <sub>2</sub> SiO <sub>3</sub> .9H <sub>2</sub> O	1.06x10 <sup>-1</sup>				
Trace n	netals				
FeCl <sub>3</sub> .6H <sub>2</sub> O	1.17x10 <sup>-2</sup>				
Na <sub>2</sub> EDTA.2H <sub>2</sub> O	1.17x10 <sup>-2</sup>				
CuSO <sub>4</sub> .5H <sub>2</sub> O	3.93x10 <sup>-5</sup>				
Na <sub>2</sub> MoO <sub>4</sub> .2H <sub>2</sub> O	2.60x10 <sup>-5</sup>				
ZnSO <sub>4</sub> .7H <sub>2</sub> O	7.65x10 <sup>-5</sup>				
CoCl <sub>2</sub> .6H <sub>2</sub> O	4.20x10 <sup>-5</sup>				
MnCl <sub>2</sub> .4H <sub>2</sub> O	9.10x10 <sup>-5</sup>				
Vitamins					
thiamineHCl (vit. B1)	2.96x10 <sup>-4</sup>				
biotin (vit. H)	2.05x10 <sup>-6</sup>				
cyanocobalamin (vit. B12)	3.69x10 <sup>-7</sup>				

**Table 2.** Modelling of *Chlorella m.* based on R<sub>c</sub> as a function of biomass concentration. Data is derived from Figure 4 and Equation 1. R<sub>m</sub> was determined as 6.18E+12 m<sup>-1</sup> and the dependence of R<sub>c</sub> on biomass concentration is given as the best fit of a natural log function  $R_c = 8.474E + 12 \times \ln(Concentration) + 6.258E + 12, r^2 = 0.889$  (Equation 5)

Biomas	SS		
concentra	stion Slope	r²	R <sub>c</sub> (m <sup>-1</sup> )
(g DCW)	/L)		5
0.52	1.185E-10	0.986	3.29E+12
1.12	1.081E-10	0.988	4.20E+12
1.52	8.082E-11	0.984	7.71E+12
2.42	5.675E-11	0.961	1.36E+13
3.14	4.586E-11	0.975	1.83E+13
4.03	4.586E-11	0.975	1.83E+13

**Table 3.**Influence of the operating parameters in the harvesting costs of *Chlorella* minutissima and *Scenedesmus species* for 600 L of culture. 1 - experimental, 2 - increase of  $\Delta P$ , 3 - increase of initial biomass concentration, 4 - increase of membrane area, 5 - increase in temperature.

Operating conditions	1	2	3	4	5	Optimal	
V (L)	600	600	600	600	600	600	
TMP (Pa)	1.95E+05	2.10E+05	1.95E+05	1.95E+05	1.95E+05	1.95E+05	
Area (m²)	3.8	3.8	3.8	7.6	3.8	7.6	
Initial concentration  (g DCW/L)	1	1	2	1	1	2	
Temperature (K)	293	293	293	293	303	283	
Chlorella minutissima							
kWh/m³	2.86	2.72	3.87	1.43	2.27	2.51	
kWh/kg	2.86	2.72	1.93	1.43	2.27	1.27	
\$/kg	0.37	0.35	0.25	0.18	0.29	0.16	
		Scenedesmu	ıs species[1]	7]			
kWh/m³	2.23	2.07	2.68	1.08	1.72	1.74	
kWh/kg	2.23	2.07	1.34	1.08	1.72	0.87	
\$/kg	0.28	0.27	0.15	0.14	0.22	0.11	
\$/kg 0.28 0.27 0.15 0.14 0.22 0.11							

**Table 4.** Influence of initial biomass concentration on harvesting costs as a function of volumetric concentration factor (VCF) considering temperature at 20 °C, 7.6 m<sup>2</sup> membrane area,  $\Delta P$  at 1.95 bar and pump efficiency of 85 %.

Initial concentration (g DCW/L)	Volume (L)	Final concentration (g DCW/L)	VCF	Energy consumption (kWh/m³)	Biomass cost (\$/kg microalgae)
0.5	2000	100.0	200.0	0.97	0.25
0.75		150.0	200.0	1.27	0.22
1.0			133.3	1.47	0.19
2.0			75.0	1.95	0.13

**Table 5.** Energy comparison of membrane based microalgae dewatering systems.

Membrane type	Scale	VCF	Maximum concentration	Microalgae species	Cost estimates	Ref.
,,			(g DCW/L)		(kWh/m³)	
MF	Pilot	150	150	Chlorella minutissima	1.3	This work
MF	Pilot	100	150	Scenedemus sp.	0.9	[17]
MF	Lab	100	89	Tetraselmissuecica	2.1	[27]
MF	Lab	75	150	Nannochloropsis sp.	0.7	[16]
MMV	Lab	15	<1.5	Phaeodactylumtricornutum and Chlorella vulgaris	0.2	[28]
FO	Lab	<20	<20	Chlorella vulgaris	0.3	[29]

#### Highlights

- Pilot-scale investigation of harvesting *Chlorella minutissima* by microfiltration.
- ACCEPTED MANUSCRIP Modelling and harvesting costs determined as a function of energy consumption.
  - Influence of the operating parameters on harvesting costs was investigated.