UNESCO-IHE
Institute for Water Education

ANII
AGENCIA NACIONAL
DE INVESTIGACIÓN
E INNOVACIÓN

BIOTHANE
Leading Anaerobic Technologies

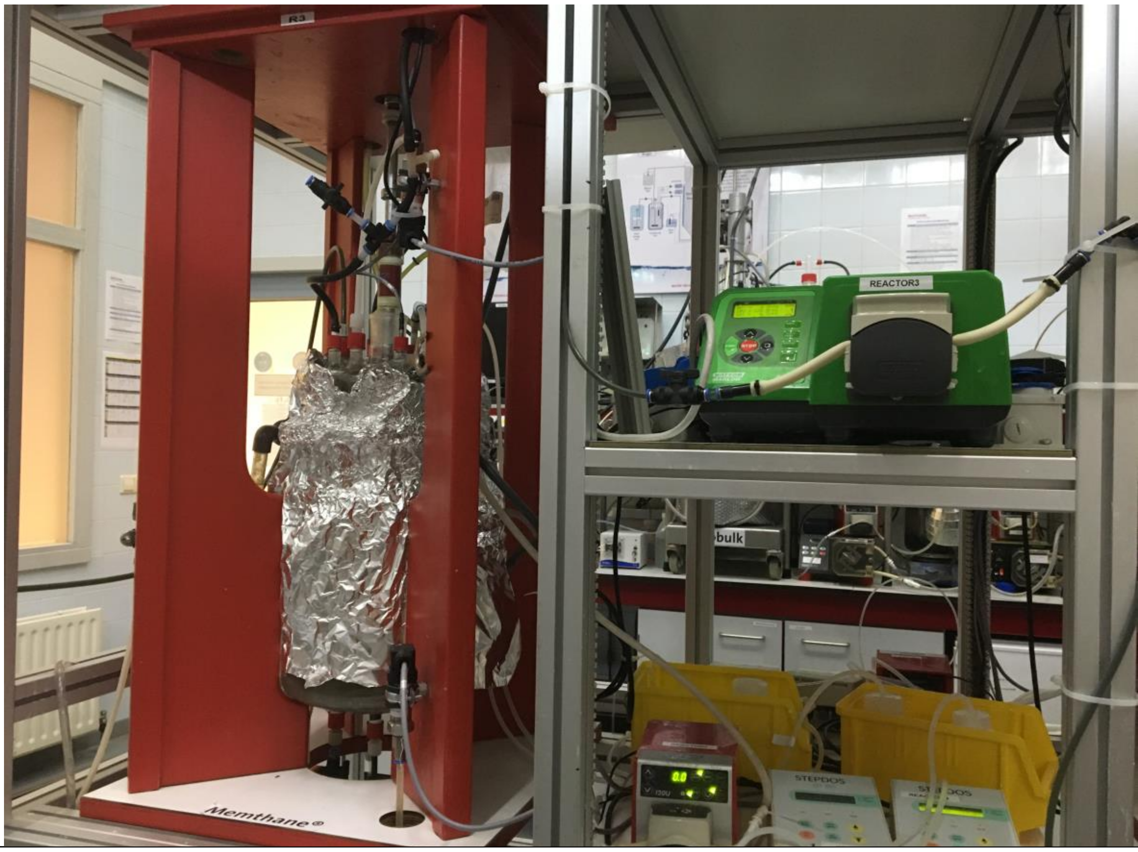
Dairy effluent treatment in an Anaerobic Membrane Bioreactor: Assessment of filtration performance

Giannina Pinotti Alonso

MSc Thesis UWS-SE.18-07

March 2018

Under embargo till March 2019



Dairy effluent treatment in an Anaerobic Membrane Bioreactor: Assessment of filtration performance

Master of Science Thesis
by
Giannina Pinotti Alonso

Supervisors
Prof. Jules van Lier

Mentors
Hector A García, Ph.D., M.Sc.
Tineke Hooijmans, Ph.D., M.Sc.

Examination committee
Prof. Jules van Lier
Hector A García, Ph.D., M.Sc.
Diana Miguez, Ph.D.- LATU

This research is done for the partial fulfilment of requirements for the Master of Science degree at the UNESCO-IHE Institute for Water Education, Delft, the Netherlands

Delft
March 2018

Abstract

Dairy industry contribution to environmental pollution is highly renowned worldwide due to its high water consumption and consequently wastewater generation. Particularly in Uruguay is responsible for a vast portion of organics and nutrients discharge to Santa Lucia river basin causing water quality problems in the area. Therefore, it is necessary to develop reliable and sustainable systems for treatment and potential reuse of the wastewater of this sector. In this context, anaerobic treatment has been widely applied, especially to highly concentrated WW, due to its benefits over aerobic treatment such as lower energy consumption, methane production, lower footprint and less waste sludge generation. However, problems related to high lipid content in dairy WW require the application of pre-treatment steps reducing the profitability of this technology.

The use of AnMBRs results of special interest for dairy WW treatment given their characteristic of full biomass retention and effluent of a generally good quality that can be used as a precursor for nutrient recovery or reuse treatment processes. However, fouling issues affecting life-span of the membranes and regular reactor operation still restrict the sustainability of AnMBR systems. Optimization of operational parameters is required in order to reduce this issues at a minimum cost, and several lab-scale studies have been done over the last decades towards this objective.

In this research, the feeding of an AnMBR that was already in operation for more than 400 days was changed from synthetic WW (diluted milk) to real dairy WW (whey and wash water). Particularly, the feast-famine regime effect on biological and filtration performance of the AnMBR was planned to be evaluated.

In terms of biological performance, for a defined VLR of 6 g COD/Lday and SRT of 30 days the average COD removal measured was 98.5% and the observed methane conversion of 0.27 L_{CH4}/kg $COD_{removed}$. Because of the low pH and alkalinity of the feed, addition of base was necessary during the first stage of the system operation to maintain the pH in the reactor around 7. Base addition was stopped trying to reduce crystals precipitation in the system. After that, the pH dropped to an equilibrium value of 6.8 without affecting the sludge activity or biological performance. However, a presumable negative effect on sludge properties leading to a higher fouling tendency was observed. Both capillary suction time and viscosity reached values around 2000 s and 16 mPas, respectively.

As far as filtration is concerned, an average flux of 12 Lmh was measured for the operation at a TMP around 400 mbar. These values were not in accordance with full-scale reported permeation rates for similar industries (20 Lmh and 100-200 mbar) leading to the necessity for a deeper evaluation of filtration performance in this system. A specific methodology for the assessment of cross-flow and cycle duration effect on permeability was used, and recommendations for its future application in other AnMBRs were made.

From the results of cross-flow velocity effect for different membrane conditions (assessed through clean water permeability tests), a linear relation between flux and cross-flow was observed within the studied range (1 – 1.6 m/s). An average increase of 7-15% in the flux was observed every 20% of the increase in cross-flow velocity. According to these results, it is not profitable to operate this system at higher cross-flow velocities than 1 m/s as the increase in

energy consumption for pumping is not compensated by the improvement in flux. Additionally, no evaluation of shear stress on sludge filterability properties was done, but a negative effect due to operation at higher cross-flows is expected in the long-term.

Regarding cycle duration, two conditions of backwash frequency were compared: every 15 (regular operation) and 30 minutes. A significant reduction in flux (>16%) for the lower frequency was measured for all the cross-flow velocities tested. Additionally, membrane CWP was meaningfully reduced during the operation at these longer cycles even considering the short-term of this experiments. Therefore, backwash frequency proved to be a relevant method for fouling control in this system.

Finally, the high suction applied through the permeate pump was identified to have a negative effect on the permeation rate obtained in the system. This is likely related to the rapid development of cake layer promoted by the negative pressure on the permeate line. Consequently, modification of the set-up to avoid suction was suggested as an improvement in order to replicate faithfully the filtration conditions of full-scale systems.

Keywords: Anaerobic membrane bioreactor; real dairy effluent; intermittent feeding; cross-flow velocity; backwash frequency

Acknowledgements

I would like to start citing the African proverb "If you want to go fast, go alone. If you want to go far, go together". This thesis would not have been possible without the support of all the people I will mention in this section. They either contributed to my work or to my personal life.

First of all, I would like to express my gratitude to my supervisor Prof. Jules van Lier and my mentors Hector García and Tineke Hooijmans for their guidance and dedication during this thesis. I am also particularly grateful for the economic support of ANII that made it possible for me to have this wonderful experience at UNESCO-IHE Delft.

I would also like to thank Biothane® for giving me the opportunity to conduct this thesis in their facilities. Additional thanks to Santiago Pacheco-Ruiz and Frans Willem Hammer that were my support on a daily basis. I am grateful to all the people I met while working in the lab for their good spirit and collaboration, including Assia, Ayunda, Chiara, Paola, Rewin, Menno and Ana. Also thanks to the people in IHE laboratory for being available when it was required and particularly to Berend Lolkema for his disposition to help with the SEM/EDS analysis. Special thanks also to Mohammed Jafar from TU Delft laboratory for being nice and patient to teach me about PSD analysis. I appreciate the collaboration also of KU Leuven university for the sludge evaluation and particularly to Pieter Van Gaelen, all the best with his PhD!

I am also deeply grateful to all my friends in Delft for constituting an additional family on this other side of the world. Especially to all my UWS class and our second mamma Yness Slokar. All my gratitude to my friends Alejandro Medina, John Koppoe, Arjun Sharma, Cristo Perez, Carolina Martinez and Carlos Lu, they all know wherever they are they will be forever in my heart. I would also like to thank Carolina Betinelli for helping with everything I needed before coming and once in Delft. A special recognition to my friend Lucía Poppolo who followed me in this crazy idea of coming to Delft and once more will share the next adventure of travelling around Asia.

Despite the distance, I don't want to forget the people in Uruguay that have always been there for me. All my family that was very supportive not only during this thesis but on all the decisions I have taken along my life. All my love and gratitude to my mom for inspiring me with her spirit, showing that anything can be achieved with effort and helping others on your way. Also to my close family, my brother Paolo, my grandfather and father, both Mario, my mother's husband Jorge and my brother's fiancée Valeria (wishing nephews soon). Not to forget the ones that are not with us anymore but were part of whom I became, my adored grandparents Deolinda, Eloides and Servando.

Finally, to all my beloved friends in Uruguay for their unconditional support through all this experience. Especially, to all the ones that made an effort to visit me during my stay in Delft: Pascua, Mamá, Jorge, Caro, Fede, Fio, Ana, Oscar, Lautaro, Pao and Vale. We really had a great time, and you helped me to shorten the distance.

"Ce n'est pas la distance qui mesure l'éloignement."

\- *Antoine de Saint-Exupéry* -

Table of Contents

List of Figures

List of Tables

Abbreviations

ABR	Anaerobic baffled reactor
AF	Anaerobic filter
AnMBR	Anaerobic membrane bioreactor
ANOVA	Analysis of variance
BMP	Biochemical methane potential
CAPEX	Capital expenditures
CAS	Conventional activated sludge
CIP	Clean in place
COD	Chemical oxygen demand
CSRT	Continuous stirred tank reactor
CST	Capillarity suction time
CWP	Clean water permeability
BOD_5	Biological oxygen demand
DINAMA	Environmental Protection Agency of Uruguay
EDS	Energy-dispersive X-ray Spectrometry
EGSB	Expanded granular sludge bed
EPS	Extracellular polymeric substances
FOG	Fats, oil & grease
HRT	Hydraulic retention time
IC	Internal circulating
JICA	Japan International Cooperation Agency
LCFA	Long-chain fatty acid
Lmh	Litre per square metre per hour
MBR	Membrane bioreactor
MF	Microfiltration
MLSS	Mixed liquor suspended solids
MVOTMA	Ministry of Housing, Land Planning and Environment
MWCO	Molecular weight cut-off
OLR	Organic loading rate
ONU	United Nations
OPEX	Operational expenditures

PAC	Powdered activated carbon
PLC	Programmable logic controller
PSD	Particle size distribution
RH	Relative hydrophobicity
SEM	Scanning electron microscopy
SKN	Soluble Kjeldahl Nitrogen
SLR	Sludge loading rate
SMA	Specific methanogenic activity
SMP	Soluble microbial products
SRT	Sludge retention time
SS	Suspended solids
TKN	Total Kjeldahl Nitrogen
TMP	Transmembrane pressure
TP	Total phosphorous
TS	Total solids
TSS	Total suspended solids
UASB	Up-flow Anaerobic Sludge Blanket
UF	Ultrafiltration
UNEP	United Nations Environment Programme
VFA	Volatile fatty acid
VLR	Volumetric loading rate
VSS	Volatile suspended solids
WW	Wastewater
WWT	Wastewater treatment

CHAPTER 1

Introduction

1.1 Background information

It is estimated that 80% of the total wastewater generated around the world is being released to the environment with improper treatment (ONU, 2015). In low and middle-income countries only 8% to 38% of the wastewater is treated (Sato et al., 2013). This causes many rivers in Africa, Asia and South America to suffer high levels of pathogens and organic pollution (UNEP, 2016). Particularly, Santa Lucia river basin, which is one of the main basins in Uruguay, has been undergoing eutrophication episodes due to agricultural runoff, sewage and poorly treated industrial wastewater discharge. This river is the source of drinking water for 60% of the country's population (JICA and MVOTMA, 2007).

Industrial establishments related to food and beverage production in the area were identified by JICA and MVOTMA (2007) as the main point-sources of contamination. Dairy industry is one of the biggest in the country, and it is the third most relevant in the Santa Lucia region (after meat processing and leather industry), representing 13% of the total organic load and 10% of nitrogen and phosphorous discharged from industrial activities in the basin (DINAMA and JICA, 2010). Currently, ponds and wetlands are the most commonly applied treatment technologies in these industries failing to meet the more strict regulations set by the Environmental Protection Agency of Uruguay (DINAMA) to protect sensitive areas of the Santa Lucia river basin. This entails the necessity of more efficient wastewater treatment technologies to meet the discharge standards (Fraga et al., 2017). Furthermore, a new approach to wastewater treatment towards WW reuse and recovery of nutrients is required in order to make it attractive and affordable for the industrial sector.

In this regard, anaerobic treatment technologies are considered attractive as they can deal with high organic loads with almost no energy consumption, low sludge generation and the benefit of methane production. Anaerobic filters and Upflow Anaerobic Sludge Bed (UASB) reactors are the most widely applied in the sector (Demirel et al., 2005). However, the success of these systems depends on retention of slow-growing biomass by decoupling hydraulic retention time (HRT) and sludge retention time (SRT) either by attachment or granulation (Dereli et al., 2012). Problems of sludge floatation and lipids accumulation have been reported for treatment of dairy wastewaters in this systems (Vidal et al., 2000).

Anaerobic membrane bioreactors (AnMBR) represent a promising technology in terms of treatment efficiency and water reuse. This technology combines the aforementioned benefits of anaerobic treatment with the advantages of membrane bioreactors (MBR) as complete biomass retention and high effluent quality (Dereli et al., 2012). Several successful applications of AnMBR to treat food processing wastewaters, mainly at lab-scale, have been reported (Dereli et al., 2012). The main drawback identified was membrane fouling as it affects productivity and reduces the lifespan of membranes resulting in higher investment costs (Lin et al., 2013). Factors as configuration (side-stream or submerged), membrane material and pore size, operational conditions and effluent characteristics have been recognised as determinants of fouling extent (Dereli et al., 2012; Jeison and van Lier, 2006). Therefore, for each effluent, in particular, these conditions must be optimized in order to make the application of the technology economically viable.

Regarding dairy effluent, few applications of AnMBR at full or laboratory scale with real dairy wastewater have been reported (Al-Malack and Aldana, 2016; Bouman and Heffernan, 2010; Dereli et al., 2012). Consequently, further studies are required for the optimization of the operational conditions in order to avoid issues caused by the high lipids content of the wastewater. Cavaleiro et al. (2008) verified in batch experiments a positive effect of intermittent feeding on the anaerobic degradation of dairy WW treatment. Further research should be conducted to define the effects of feeding strategy on dairy wastewater in AnMBR and the optimal operational conditions to make viable the application of this technology.

1.2 Problem statement

The dairy industry is responsible for a big percentage of pollution worldwide as it uses great amounts of fresh water (Vourch et al., 2008). AnMBR is a promising technology for dairy wastewater treatment and reuses due to its high effluent quality and methane production. However, despite the decline in membrane costs during last decades, they still represent the biggest portion of capital costs for this systems (Dereli et al., 2012). Optimization of operational conditions to reduce fouling and increase membrane flux can directly contribute to the economic viability of this technology.

Synthetic dairy wastewater (diluted) was treated in a side-stream bench-scale AnMBR in Veolia-Biothane as part of the PhD research of Alejandra Szabo. However, after the three SRT's, the filterability of the sludge measured decreased to a level at which it was not feasible to operate due to the high trans-membrane pressure (TMP) required. The capillary suction time (CST), used as an indicator of sludge quality increased up to 1500 s during this period. After the change to sequential operation, it dropped to 400 s and was maintained at values lower than 800 s the next 180 days of operation. However, flux values remained around 10 – 12 Lmh independently of sludge conditions.

Considering that some existent full-scale installations in the dairy sector operate at around 20-25 Lmh with TMPs of 100-200 mbar (Bouman and Heffernan, 2010) with sludge of higher CST values, lab-scale results for membrane performance evaluation need to be treated with caution. Additionally, the type of substrate has a critical role in membrane fouling (Dereli et al., 2012; Le-Clech et al., 2006) and it is, therefore, important to evaluate the effect of treating real instead of synthetic WW. Furthermore, it is desirable to verify the effect of the feast-famine regime before its application in full-scale plants as it would increase capital costs of the installation (e.g., buffer tanks, and feeding control required). Considering the aforementioned factors, counting with a lab-scale system that allows both biological and membrane performance evaluation as close as possible to full-scale conditions is necessary for decision-making on full-scale designs.

1.3 Research questions

In the context of the literature review performed and the interest of application of AnMBR at full-scale for the treatment of dairy wastewater, the following research questions arose.

1- How is the biological and filtration performance of a laboratory AnMBR operated in a feast-famine regime using real dairy wastewater?
2- Which are the main characteristics of the sludge developed in a laboratory AnMBR operated in a feast-famine regime using real dairy wastewater? Which of them are better related to membrane performance?
3- In how far can the laboratory results be extrapolated for full-scale systems?
4- Which of the membrane operational parameters (backwash frequency or cross-flow velocity) have the highest effect on the permeability and flux of the selected system?

1.4 Goal and Objectives

The main goal of this research is to evaluate the performance (biological and filtration) of a bench-scale AnMBR treating real dairy wastewater in feast-famine regime in order to contribute to the future design of a full-scale system.

Specific objectives of the research were defined, as follows.

1- Characterize real dairy processing wastewater in order to relate the AnMBR operational performance with its particular characteristics.
2- Evaluate the biological performance (methane production, volatile fatty acids (VFA) concentrations and chemical oxygen demand (COD) removal) of the AnMBR.
3- Evaluate the sludge filterability properties (CST, viscosity, morphology and particle size) and membrane permeability of the AnMBR.
4- Determine the optimum combination in terms of flux and permeability of backwash frequency and cross-flow velocity of the AnMBR.
5- Establish a standard methodology for the evaluation of optimum cross-flow velocity and backwash frequency at bench-scale studies.
6- Propose any modifications necessary for the bench-scale set-up in order to extrapolate the results to full-scale operational values.

CHAPTER 2

Literature Review

In this chapter, relevant information regarding the research topic and a critical review of it will be presented. Firstly, a description of the dairy sector in general and specifically its environmental effects in Uruguay are shown. Secondly, literature data about characteristics of dairy wastewater in terms of anaerobic treatment is summarized, and possible effects on the anaerobic treatment are assessed. Thirdly, anaerobic treatment principles and technologies are discussed, especially focusing on dairy wastewater application and intermittent feeding cases. Finally, AnMBR wastewater treatment principles and main issues encountered are presented including operational parameters effect on reactor performance and application examples on industrial WW treatment.

2.1 Dairy industry sector

The dairy industry is one of the most important ones in the food production sector, corresponding to an average of 14% of the total agricultural products marketed worldwide, reaching up to 20% for some countries. In the next 10 years, the sector is forecasted to grow at a yearly rate of 1.8% (Food and Agriculture Organization of the United Nations, 2016). According to Vourch et al. (2008), the dairy industry is one of the biggest polluters in terms of the high volume of wastewater generated. The heterogeneity of the processes in this sector results in a wide range of water volume consumed per litre of processed milk. Most of the industries consume between 1 to 10 litres of water per litre of processed milk being the wastewater production directly related to that value (Wojdalski et al., 2013).

2.1.1 The situation of the sector in Uruguay

Uruguayan's economy mainly relies on agro-industrial activities. In 2016, 78% of the total goods exported corresponded to agro-industrial products. The dairy industry is one of the main ones in the sector accounting for 8% of the total exports in 2014. The activity has been steadily increasing in the last 10 years at an average annual rate of 2.6% (Uruguay_XXI, 2015).

However, environmental challenges for this and other food industries, related to wastewater disposal, reuse and recovery have arisen in the last decade due to severe cases of rivers pollution. The most relevant example is the one of the Santa Lucia river basin (Figure 2-1) as it represents the source of freshwater for around 60% of the country's population, including the capital city Montevideo (JICA and MVOTMA, 2007). Algae bloom issues in the river due to excessive nutrients discharged from industrial facilities, sewage and agricultural runoff have affected the performance of the drinking water treatment plant resulting in episodes of bad odour and taste in the supplied water.

Currently, 7 dairy industries are in operation in the Santa Lucia river basin area. From all the industrial activities in the region, this sector discharges 13% of the organic load and 10% of the total nutrients, mainly nitrogen and phosphorous. Additionally, 40% of these dairy production facilities do not comply with the standard of biological oxygen demand (DBO_5) for discharge to a river (60 mg/L) and almost all the facilities exceed the 10 mgTKN/L and the 5 mgP/L stated in the national decree 253/79 (Japan International Cooperation Agency and Ministry of Housing Land Planning and Environment, 2011).

Considering that footprint is not an issue in most of the areas where the industrial facilities are located, stabilization ponds and wetlands were the preferred systems due to their low operation and maintenance cost. However, these systems cannot comply with the stricter standards established for the preservation of the Santa Lucia basin, especially for nutrients removal. Therefore, it is necessary for the companies to invest in more efficient treatment technologies (Fraga et al., 2017). Alternatives that allow recovery of resources and reuse of treated wastewater need to be prioritized in order to make the investment attractive for the industries.

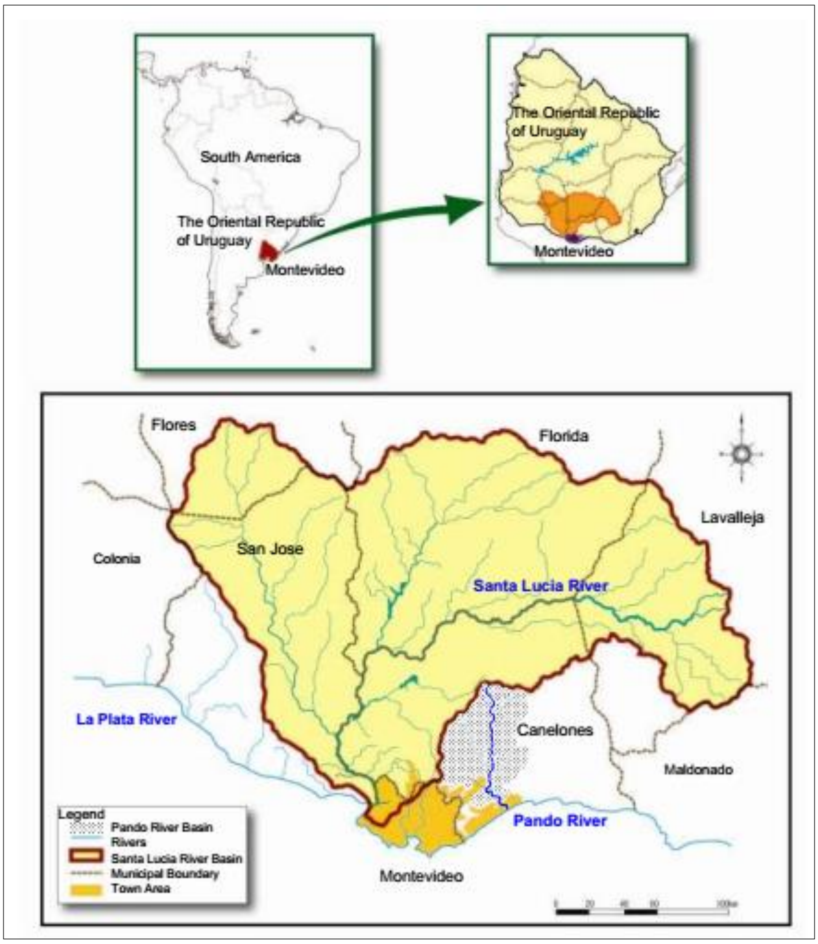


Figure 2-1 Uruguay and Santa Lucia river basin location. Source: Japan International Cooperation Agency and Ministry of Housing Land Planning and Environment (2008)

2.2 Dairy wastewater

Dairy wastewater generation, including out of specification product discharge, is estimated in average as 2.5 litres per litre of processed milk. It is mainly produced during cleaning of storage tanks, process equipment and transport lines between cycles, so the amount increases when different products are produced on the same line. Additionally, a wide variety of goods including yoghurt, milk, butter, cheese, desserts and powdered milk, are manufactured resulting in a wider range of wastewater volumes and concentration (Danalewich et al., 1998; Shete and Shinkar, 2013). Seasonal variations might also affect wastewater production, due to variations in the amount of milk received which tends to be higher during summer (Kolarski and Nyhuis, 1996).

Regarding its composition, dairy wastewater is concentrated in carbohydrates, proteins and fats. Generally, COD varies between 4 to 15 g/L, fats between 0.7 to 2.9 g/L and suspended solids (SS) from 0.2 to 5.1 g/L (Danalewich et al., 1998; Demirel et al., 2005; Shete and Shinkar, 2013). The main carbohydrate is lactose which is easily biodegradable by anaerobic bacteria. High levels of lactose were reported to have a negative effect on proteolytic production affecting protein degradation (Fang, 2000). However, in case of adapted sludge, casein, which is the main protein in the dairy effluent, can be rapidly anaerobically degraded (Perle et al., 1995).

Lipids degradation in the wastewater produces long chain fatty acids (LCFAs) that are known to be the main inhibitory factor in the anaerobic degradation of lipid-containing wastewater (Hanaki et al., 1981). The effect is mainly related to adherence to the biomass affecting the mass transfer (Hwu et al., 1997). Also, biomass floatation and wash-out issues related to LCFAs have been reported in granular anaerobic reactors (Hwu et al., 1997; Vidal et al., 2000). Nevertheless, the inhibitory effect is reported to be reversible, recovering the methanogenic capacity after depletion of LCFAs (Pereira et al., 2003).

A particular flow that might end up in wastewater in the dairy industry, unless especially treated or recovered as animal feeding, is cheese whey. It is mainly composed of lactose as most of proteins and lipids are coagulated in the cheese making process. Therefore, degradation of lactose leads to rapid formation of VFAs when anaerobically treated (Gutiérrez et al., 1991). Also ,its acidic composition (pH normally lower than 5), high COD concentrations (60–100 g/L) and relevant salinity (~8 μS/cm) is challenging for anaerobic processing (Carvalho et al., 2013). Although cheese whey mixed with wastewater has been treated in anaerobic systems in the last 20 years (Carvalho et al., 2013), VFAs building up in the systems has affected methane production (Rodgers et al., 2004) and granule formation (Yang et al., 2003). Hence, special considerations for the anaerobic treatment processes should be taken in case cheese whey WW is discharged to the wastewater treatment plant.

2.3 Anaerobic treatment

Anaerobic processes had its first enforcement in 1860 in France by Louis M. Mouras and Abbé Moigno for treatment of sewage (van Lier et al., 2008). From that point on, despite that, it was widely applied in cities and benefits of biogas generation recognized, not many improvements in understanding of the process microbiology and biochemistry were made. More knowledge about aerobic processes was available being preferred in the 30's due to their efficiency. Just 50 years ago the anaerobic treatment regained attention due to the progress of anaerobic filters in the 60's powered by Perry McCarty from the US and the novelty of UASB reactors introduced in the Netherlands by Gatze Lettinga (Lettinga, 2010).This set the start point for the wide application and development of high rate anaerobic technologies such as expanded granular sludge bed (EGSB), internal circulating (IC) and anaerobic baffled reactors (ABR). All these technologies, particularly the UASB reactors have been broadly applied in the treatment of wastewaters, especially for highly concentrated streams from agro-industries (Lettinga, 2010).

The number of installations shows an exponential growth of constructions which is directly related to the important benefits that this treatment has compared to aerobic processes. In 2007 more than 2226 operational anaerobic reactors were registered worldwide and around 500 more with no license from any company (van Lier et al., 2008). In general, the main advantage can be seen in the energy balance (Figure 2-2) as no energy for aeration is required and approximately 13.5 MJ as CH_4/kg COD removed is produced (1.5 kWh with 40% efficiency). Additionally, high savings on sludge treatment and disposal are made as the volume is up to 10 times lower, as shown in Figure 2-2, than in conventional activated sludge systems (CAS).

Other benefits from anaerobic treatment can be enumerated, and its significance will be finally determined by application conditions. Some of them are: smaller footprint (up to 90% smaller in granular bed technologies), high loading rates can be achieved (20-35 kgCOD/m^3d), market value of granular sludge, rapid start-up when inoculated with granular sludge, lower nutrient requirements and possibility of interrupting operation for months without affecting the biomass which makes it suitable for seasonal processes (Switzenbaum, 1983; van Lier et al., 2008).

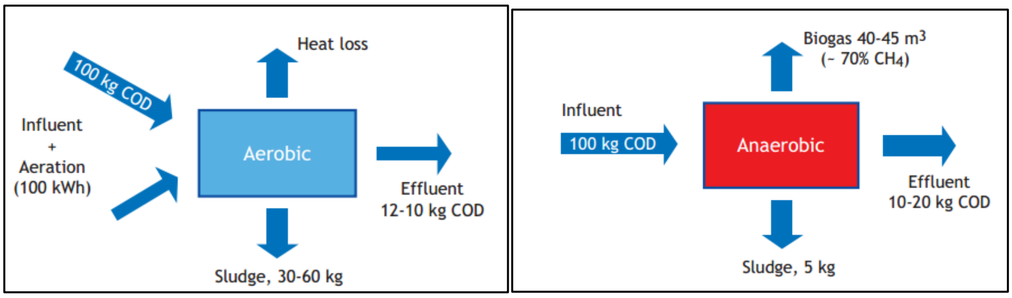


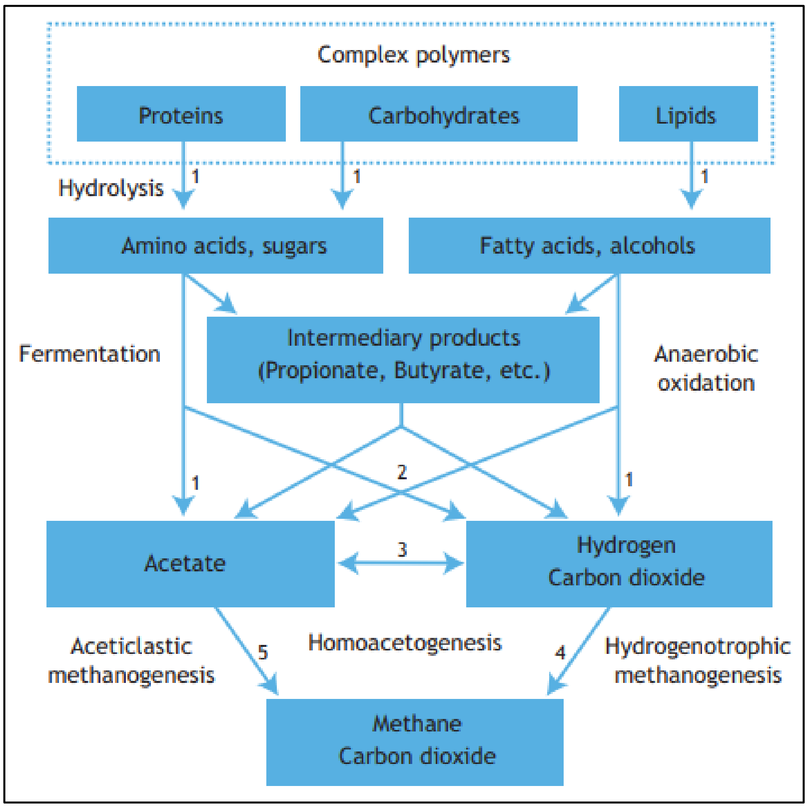
Figure 2-2 Mass and energy flows in aerobic and anaerobic degradation. Source: van Lier et al. (2008)

However, there are still some drawbacks of anaerobic systems compared to aerobic treatment. The main disadvantage is related to effluent quality. Particularly, the higher concentrations of COD (Figure 2-2) and nutrients in the effluent (Nadais et al., 2010) leading to the necessity of additional post-treatment steps before discharge. Additionally, higher operational temperatures, bigger volumes requirements due to a slower degradation rate and worse stability of the process (Switzenbaum, 1983) are still factors that require improvement in the anaerobic treatment.

2.3.1 Microbiology of the process

In anaerobic digestion, consortia of bacteria and archaea degrade the organic matter in four steps transforming it into methane (CH_4), carbon dioxide (CO_2), ammonium (NH_3), hydrogen sulphide (H_2S) and water (H_2O) as end-products. Figure 2-3 summarizes the compounds, groups of bacteria and archaea involved, processes and their interactions.

The first step is ***hydrolysis***, were mainly fermentative bacteria segregate enzymes to break-down undissolved polymers into dissolved smaller molecules that can enter the cell through the membrane. Also, other microorganisms were related to this first step as protozoa, fungi and yeasts. The products of that step are used by the fermentative bacteria for cell growth in the ***acidogenesis*** where simple compounds are generated as by-products (VFAs, lactic acid, alcohols, CO_2, H_2, NH_3 and H_2S). Afterwards, in the ***acetogenesis***, VFAs are transformed into acetate, H_2 and CO_2. The last step is the ***methanogenesis***, in which archaea species produce methane and carbon dioxide from acetate (acetoclastic methanogens) or from H_2 and CO_2 (hydrogenotrophic methanogens) (Novaes, 1986).



1- Hydrolytic and fermentative bacteria; 2- Acetogenic bacteria; 3-Homo-acetogenic bacteria; 4-Hydrogenotrophic methanogens; 5-Aceticlastic methanogens.

Figure 2-3 Scheme of anaerobic digestion processes. Source: van Lier et al. (2008)

In anaerobic wastewater treatment, methanogenesis is normally the bottle-neck, especially during start-up, as methanogens have the lowest growth rate among these groups of microorganisms (E. Griffin et al., 1998). Due to this reason is that, unless there is adapted biomass available as seed, the start-up of high rate reactors can take several months (van Lier et al., 2008).

Additionally, these archaea are very sensitive to pH drops so special attention needs to be paid to avoid overloading of the methanogenic capacity of the systems. In that case, accumulation of high amount of VFAs causes pH drops which directly affect the methanogenic activity. Batch operation of the reactor is required until VFAs are consumed and pH recovered (van Lier et al., 2008). In case of a high pH drop, the addition of base is a common practice (McCarty, 1964). Other components that may inhibit the methanogenic activity and therefore should be considered for reactors start-up and operation are LCFAs (see section 2.2) and ammonia.

2.3.2 Established technologies

As mentioned in section 2.3, anaerobic treatment has been widely applied in the last decades due to the development of effective high rate systems that rely on the separation of SRT and HRT. This is necessary to allow development of slowly growing biomass (methanogens) and at the same time, economic viability of the treatment using small reactors. The main methods that have been applied with that purpose are attachment and granulation.

Fixed film reactors are based on the attachment which consists in the immobilisation of the biomass in a biofilm that grows over a solid support. Different configurations of fixed film reactors as anaerobic filters and anaerobic expanded bed reactors were developed with also varying flow direction and support material (Switzenbaum, 1983). Due to the high biomass retention promoted by its attachment to carriers, shorter start-up phases and higher loading rates can be applied in these reactors (Karadag et al., 2015a). This lead to several applications of this technology during 80's and 90's (van Lier et al., 2008).

On the other hand, clogging issues in the beds, caused mainly by high solids and fats content in the wastewater, have led to short-circuiting issues and therefore loss of COD removal efficiency (Karadag et al., 2015a). In relation with these problems, the number of installations has decreased in the last decades, representing only 1% of the new reactors installed between 2002 and 2007 (van Lier et al., 2008).

Granulation based reactors represent 90% of the total anaerobic reactors installed. The main ones are UASB, EGSB and IC from which UASB represents 50% of reactors constructed between 1981 and 2007 (van Lier et al., 2008). The UASB reactor's main innovative feature is the efficient separation of biomass, water and biogas in a tank, based on granules formation and a three-phase separator installed in the top part of the reactor. There, the biogas is collected in an inverted cone after passing through a series of baffles that retain the sludge to the reactor and divert liquid flow out (Lettinga et al., 1980). Additionally, biomass-substrate contact is achieved by the up-flow of evenly distributed effluent at the bottom of the tank and the biogas produced, saving mixing energy (Daud et al., 2018).

An upgrade of UASB reactors' loading rates was achieved by expanded bed systems as EGSB and IC. In those technologies, high velocities and recirculation are applied to enhance biomass and substrate contact (Lettinga et al., 1997). That allows full use of the reactors biomass, avoiding inerts' build-up and enhancing the performance based on more efficient use of the reactors' volume. Due to their characteristics, these systems allow treatment of special wastewaters that affect UASB operation, for example, those leading to foam formation or LCFAs accumulation (van Lier et al., 2008).

Despite the high number of applications of the aforementioned technologies, there are some industrial wastewater characteristics that disturb the granules formation and therefore lead to process failures due to biomass wash-out. Some of them are related to high concentrations of compounds as fats, oil and grease (FOG), salts, toxics and suspended solids. Other cases are more related to heterogeneous effluent generation causing extreme variations in organic loading rate (OLR) and HRT of the systems. For those cases, AnMBR systems introduce membrane separation concept in order to retain the slow-growing biomass without the necessity of granules formation (Dereli et al., 2012).

2.3.3 Anaerobic treatment of dairy effluent

The high organic strength of dairy wastewater makes anaerobic treatment a preferred technology over aerobic processes due to benefits mentioned in section 2.3. High-rate treatment in both biofilm and granular reactors has been applied (Demirel et al., 2005). Biofilm reactors present benefits such as their ability to cope with loading variations and short start-up periods due to the enhanced biomass growth with surface attachment. Anaerobic filters have been the preferred option in this group. However, high suspended solids content and fats accumulation tend to cause clogging and dead zones in this reactors reducing the treatment efficiency (Karadag et al., 2015a).

Among granular reactors, UASB system has the biggest number of applications because its operation is simple and very well-known as well as the various available suppliers of this technology resulting in an easy and prompt construction process (Karadag et al., 2015b). Nonetheless, several issues due to the high amounts of FOGs have been reported. Fats accumulation and scum formation below the biogas separator produces issues in operation as mass-transfer limitations between substrate and biomass, sludge floatation, biomass loss and blockage of the three-phase separator (Passeggi et al., 2012). In order to overcome these problems, some alternatives reported are: hybrid systems combining film and granular processes (Karadag et al., 2015b); FOGs prior separation with dissolved air floatation (Campos et al., 2004); Fenton oxidation as pre-treatment (Yu and Fang, 2001) and intermittent operation strategies (Nadais et al., 2008).

2.3.4 Intermittent feeding in anaerobic treatment of dairy wastewater

As it was mentioned in section 2.3.3, dairy wastewater presents several issues for anaerobic treatment, especially related to the high lipids content. The first products of lipids degradation are long chain fatty acids (LCFAs) which tend to be adsorbed onto the sludge surface due to their high hydrophobicity. Pereira et al. (2003) studied the effect of LCFAs in an anaerobic filter concluding that the operation of the reactor in cycles composed by feeding and non-feeding periods leads to an enhancement of methanogenic activity. During the feeding period, LCFAs are accumulated on the biomass hindering methane production, and in the following stage methane is produced from their degradation (Pereira et al., 2003; Pereira et al., 2005).

Experiences of Nadais et al. (2008) in UASB reactors treating synthetic dairy wastewater confirmed previous results as 16% higher total methane production was observed in the intermittent reactor compared to the continuous feeding one. This shows that higher degradation of wastewater was achieved as a consequence of the pulse feeding regime. Similar studies but using real dairy effluent were performed by Cavaleiro et al. (2008) verifying an increase in methanation and degradation due to intermittent feeding in batch assays.

The necessity of optimizing the frequency and duration of pulses was expressed by Cavaleiro et al. (2008). Nadais et al. (2005) conducted a research with five different combinations of feeding and feedless durations concluding that the optimal for dairy wastewater was 48h:48h. Nonetheless, only equal duration of periods was compared while a longer non-feeding period than the feeding one was suggested as beneficial by Couras et al. (2015). The benefit of non-feeding periods is related to the driven adaptation of biomass, supported by changes in the microbial community, to degrade more complex substrates given that easily degradable ones are mostly consumed during the feeding stage (Nadais et al., 2006; Nadais et al., 2005). Therefore, longer fasting phases might improve that adaptation.

On the other hand, if the equal duration of the phases is pursuit, too long feeding periods might encounter the regular overloading issues of reactors as inhibition and sludge floatation. Hence, the process can be optimized by operation at longer feed-less phases. Coelho et al. (2007) operated two UASB reactors treating dairy wastewater at a combination of feeding:non-feeding periods of 6:6 and 3:9 with three different phases of same OLR applied to both reactors. The experiment showed the expected positive effect of the longer stabilization stage increasing the methanation proportion in between 4 to 6% for all the different OLR phases.

No previous studies on the effect of intermittent feeding on the operation of an AnMBR have been found. For this technology, both the effect on biological performance and filterability conditions need to be assessed. A positive influence on sludge filterability properties is expected due to a reduction of LCFA accumulation on the biomass surface.

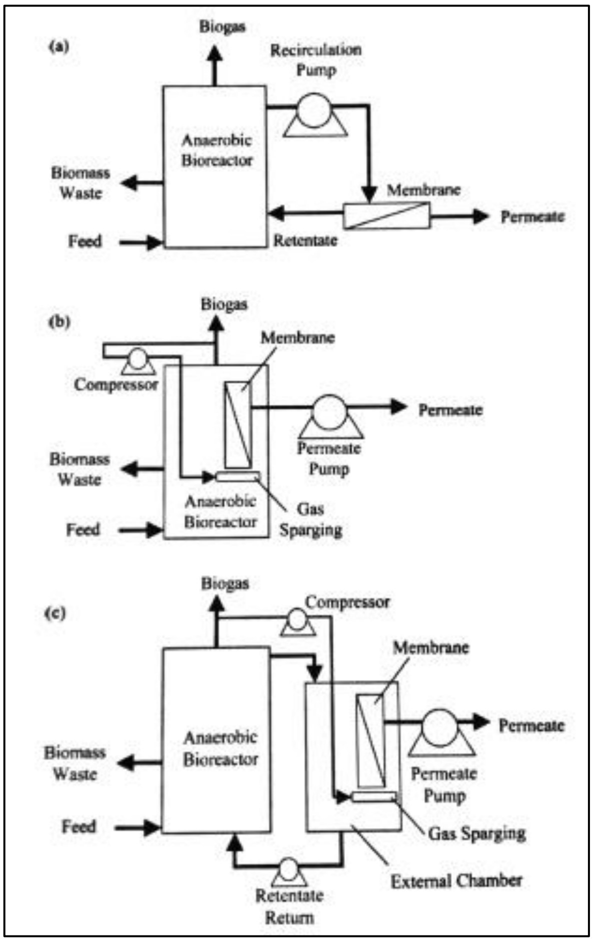
2.4 Anaerobic membrane bioreactors

AnMBRs were introduced following the idea of the MBRs of lower footprint technologies by combining solids separation and reaction in the same unit. Despite the fact that high rate anaerobic reactors could already face that challenge, some specific characteristics of wastewater as high SS content, high salinity, toxics and inhibitors concentration and FOGs produce problems both in fixed film and granular reactors (Dereli et al., 2012). Lately, due to the decreasing membrane prices, as well as the nutrient recovery and water reuse focus on wastewater treatment, the application of AnMBR became more feasible and interesting.

The idea of combining membranes with anaerobic processes was firstly introduced in a septic tank (Grethlein, 1978). Hereafter other trials of this combination were made, but they failed because of high membrane prices. The Aqua Renaissance ’90 was a program established in Japan in the nineties that tested different configurations of AnMBR for wastewater treatment reaching above 90% of COD removals (Kimura, 1991). A combination of an anaerobic digestion system with an ultrafiltration membrane was tested by Ross et al. in South Africa, 1992, achieving OLRs up to 10 kgCOD/m^{3}d, signifying the first substantial technology development in this area (Stuckey, 2010). Subsequently, in the last 25 years, several research studies especially focused on fouling as it main drawback have been done.

2.4.1 Configurations

Three different configurations can be applied in AnMBR regarding the location of the membrane (see Figure 2-4) and for all of them, advantages and disadvantages can be identified. The external position of the membrane (Figure 2-4a) simplifies operation and maintenance of the system considering membrane cleaning and replacement. However, it presents some drawbacks as energy requirement for external recirculation and decrease in floc particles size due to shear forces applied (Stuckey, 2010). For aerobic MBRs, it was concluded that centrifugal compared to rotary pumps have almost no negative effects on the biomass (Kim et al., 2001). On the other hand, methanogenic inhibition related to shear effects has been reported (Brockmann and Seyfried, 1996).



(a) Pressure-driven external cross-flow (b) Vacuum-driven submerged membrane in the reactor
(c) Vacuum-driven submerged membrane in external chamber

Figure 2-4 Membrane bioreactors configuration. Source: Liao et al. (2006)

Configurations with submerged membranes (Figures 2-4b and 2-4c) are the most applied in aerobic MBRs. The construction of an external submersion tank is used to simplify the cleaning and substitution of the modules although it requires some pumping energy. Nonetheless, these configurations are not so common in case of anaerobic MBRs. This might be related to the fact that biogas needs to be recirculated for membrane cleaning while in aerobic reactors this is done with part of the aeration supplied. Main advantages of submerged membranes are lower capital and operational costs due to energy savings and fewer pumps required (Stuckey, 2010).

2.4.2 Membrane characteristics

In general, polymer membranes (polysulfone, polyvinylidene fluoride, polyethylene, and polypropylene) are preferred due to their low cost and good resistance to chemical and thermal stress. However, they are reported to be more susceptible to fouling due to their hydrophobicity. For that reason, many studies of membrane coating have been done to combine hydrophilic compounds with these membranes. Additionally, inorganic membranes are available which tend to be more susceptible to inorganic fouling formation (Stuckey, 2010).

Regarding pore size, for most of the membranes used in bioreactors, they are between 0.02-0.5 μm (ultrafiltration (UF) to microfiltration). An optimal pore size range was identified by some studies. Big pore sizes seem to be more prone to fouling due to the accumulation of colloidal material in the pores. However, after the long-term operation, cake layers are formed over the membrane surface making the membrane properties less relevant in the filtration rates (Jeison and van Lier, 2006). This was verified by the experiments of He et al. (2005) where molecular weight cut-off (MWCO) from 20 to 70 kDa were tested, and no significant differences were found in COD, SS and bacteria removal efficiencies.

2.4.3 Fouling issues

Different factors such as reactor operational parameters, wastewater and membrane characteristics cause accumulation of inorganic and organic material covering the membrane pores in what is called fouling (Figure 2-5). This is the main problem encountered in AnMBR as it directly affects the capital costs, increasing the required membrane area in the installations due to the reduction of the flux and lifespan of membranes (Dereli et al., 2012).

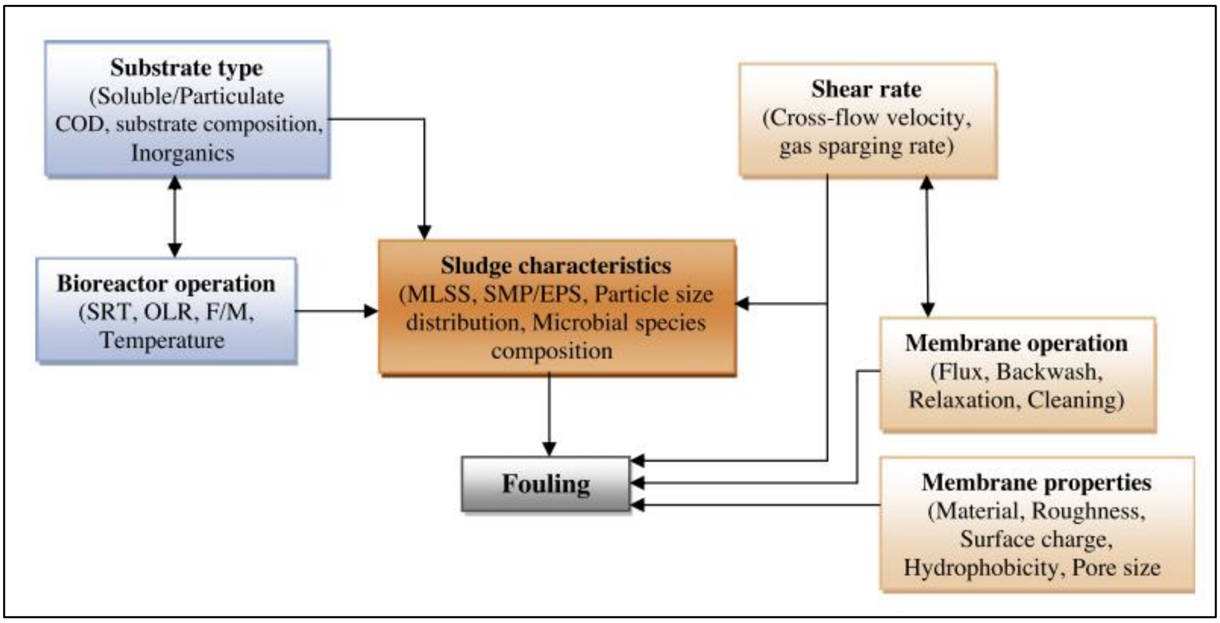


Figure 2-5 Factors influencing membrane fouling. Source: Dereli et al. (2012)

Meng et al. (2009) identified three different kinds of fouling: removable, irremovable and irreversible (see Figure 2-6). Both removable and irremovable fouling can be eliminated. For removable fouling, physical cleaning methods such as backwash or relaxation are enough while for the second a chemical cleaning process needs to be applied. The irreversible fouling still remains after chemical cleaning. As a whole, the removable fouling is associated with cake layer build-up and the irremovable with pore blocking (Meng et al., 2009). Equation 2-1 presents the calculation of the resistance according to the classification presented by Meng et al. (2009).

Equation 2-1 Calculation of membrane resistances. Source: Meng et al. (2009)

$$R_{total} = \frac{\Delta P_T}{\nu \cdot J} = R_{intrinsic} + R_{removable} + R_{irreversible} + R_{irrecoverable}$$

where:

R_{total} (m^{-1}): Filtration resistance;

ΔP_T(Pa): TMP;

ν (Pas): Kinematic viscosity of water. 8.0 x 10^{-7} Pas @ 30ºC (Crittenden et al., 2012);

J($m^3m^{-2}s^{-1}$): flux;

$R_{intrinsic}$(m^{-1}): intrinsic resistance of the membrane;

$R_{removable}$(m^{-1}): cake layer removable by flashing with water

$R_{irreversible}$(m^{-1}): resistance caused by organics and inorganics foulants and can be removed by chemical cleaning; and

$R_{irrecoverable}$(m^{-1}): resistance caused by irremovable foulants.

Fouling components can be classified as soluble organics, colloidal particles and inorganics. Soluble compounds are mainly constituted by soluble microbial products (SMPs) and extracellular polysaccharides (ECP). SMPs are products of cell lysis and metabolism, and therefore their production is increased either by imposing stressing conditions to the sludge (e.g. load or temperature shocks and toxic compounds) or by increasing SRT which encounters higher endogenous respiration activity (Dereli et al., 2014; Stuckey, 2010). Also, high lipid contents in wastewater were reported to have an effect on the amount of SMPs produced and consequently on the membrane permeability (Dereli et al., 2015).

Pore blocking is produced by colloids close to the pore size of the membranes. The shear due to pumping of the sludge increases the number of fine colloids promoting this fouling mechanism. In order to decrease the colloids in the solution, addition of powdered activated carbon (PAC) and other coagulants and flocculants, compounds has been successfully applied (Dereli et al., 2012; Stuckey, 2010).

The main inorganic foulant in AnMBR is struvite, but also other calcium or phosphate salts might be present. The relevance of these precipitants in AnMBRs is greater than in the aerobic ones. This is caused by the higher concentration of these inorganic ions in the industrial WW and also the greater availability of carbonate in the solution due to carbon dioxide equilibrium in anaerobic conditions. An additional issue of inorganic compounds precipitation is that they tend to couple with organic foulants increasing the stability of the cake layer (Dereli et al., 2012; Stuckey, 2010).

According to Stuckey (2010), three different operating strategies can be used to control fouling:

- short periods of high flux followed by relaxing/backflushing and chemical cleaning;
- flux set below critical values, so only relaxing/backflushing and eventual chemical cleaning are needed; and
- minimization of fouling by operating the reactor in order to improve sludge filterability and avoid cake layer formation.

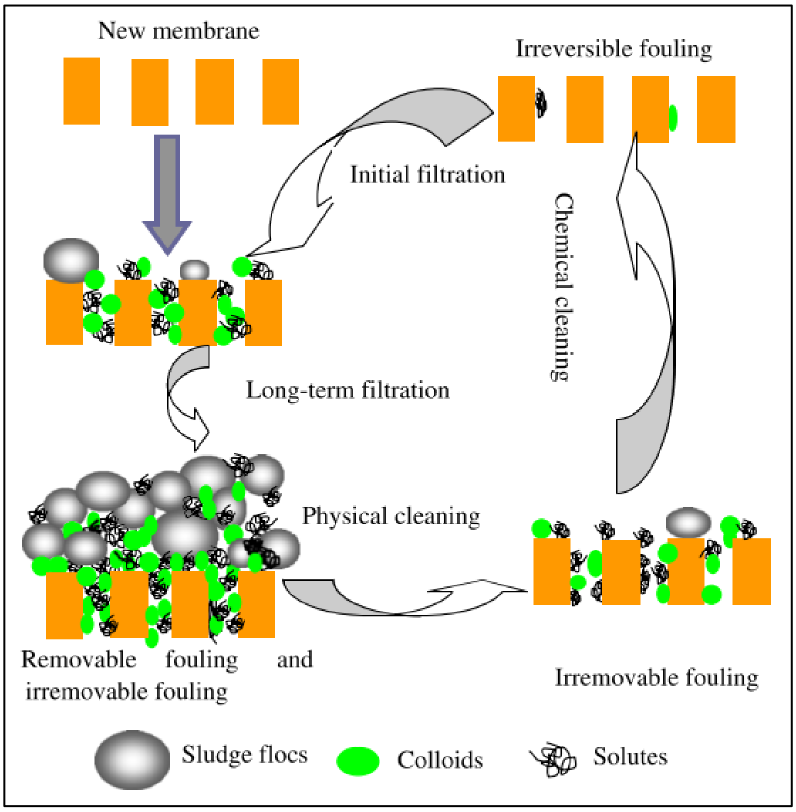


Figure 2-6 Formation and removal of fouling. Source: Meng et al. (2009)

Some examples of the last alternative mentioned can be a reduction of causes of SMP and EPS production by managing SRT, shock loads and toxics concentration; select side-stream configurations with high cross-flow velocities; and increase shear of gas on submerged membranes (Stuckey, 2010).

2.4.4 Reactor operational parameters

The following operational parameters are known to affect both biological performance and filtration efficiency.

Temperature

Increasing the temperature is expected to have a positive effect on filterability due to lower viscosity of the liquids which is a real immediate consequence observed by Jeison and van Lier (2006). Additionally, for substrates were hydrolysis is one of the limiting steps (high FOG content) operating at thermophilic conditions improves the reaction rates. However, in long-term operation, the effect is reversed as Jeison and van Lier (2007) found mesophilic fluxes to be more than two times higher than for thermophilic conditions. This result was related by the authors to the presence of smaller particles (due to the decay of mesophilic biomass from the inoculum) resulting in a more compact cake layer in the thermophilic system than in the mesophilic one. Also due to high temperatures, bacterial decay is increased and consequently SMPs and EPS as well, which are considered foulants as mentioned in section 2.4.3.

HRT

Due to the high capacity of sludge retention in AnMBRs, they are suitable to resist HRT shocks without compromising the biomass. Additionally, efficient treatment of WW at HRTs as low as 3 hours have been reported for diluted wastewater (Stuckey, 2010). However, it should be considered that low HRTs require higher biomass concentrations in the reactor and shock flows stress the biomass, both events contributing to membrane fouling (Lin et al., 2013).

SRT

As membranes allow for almost full biomass retention, the SRT can be easily controlled by selecting wastage from the reactor. In general, AnMBRs are run at SRTs in the range of 30 to 300 days (Dereli et al., 2012). Operating at high SRTs is beneficial in terms of wastage sludge post-treatment as fewer amounts of and more stabilized sludge is produced, but it has a negative effect on membrane fouling. The causes of this are the higher concentrations of biomass, and more EPS and SMPs produced due to cell decay. A negative linear correlation between stabilized flux and suspended solids concentration was found by Beaubien et al. (1996). The experiments showed an 86% decline in flux when increasing almost 10 times the TSS concentration, from 2.5 g/L to 22 g/L. Additionally, high SRT leads to higher ions concentration which might increase the inorganic fouling (Dereli et al., 2012; Dereli et al., 2014; Stuckey, 2010).

OLR

Considering biomass retention capacity of AnMBRs leading to high solids concentrations, operation at high OLRs is possible. However, the limiting factor is the activity of the sludge as for example, operating at high SRTs, high sludge concentration might not be related to active biomass available. Depending on the substrate, OLRs applied vary from 1 – 24 $kgCOD/m^3d$ (Dereli et al., 2012; Lin et al., 2013). Possibilities of increasing OLR in operating reactors should be assessed according to SMA tests in order to avoid VFAs accumulation and inhibition which results in reactor failure.

2.4.5 Membrane operational parameters

Critical flux

This characteristic of a membrane filtration system was defined by Field et al. (1995) as “a flux below which, a decline of flux with time does not occur”. However, operation in those conditions, assuming no fouling development and TMP with the same values as for clean water filtration is not appropriate for the wastewater treatment applications. Therefore, what is referred as a “weak” definition of critical flux was introduced later as the threshold value above which the relation of TMP with flux is not linear (Wu et al., 1999).

A non-linear relation of those parameters would actually translate into a variation in the permeability of the membrane. Based on this concept alternative methodologies based on fixed increments of flux or TMP have been considered for the determination of this value that can be used as a reference for system’s operation at a controlled level of fouling (Le Clech et al., 2003). Although fouling is not avoided in MBRs by operation even at low fluxes, its rate of development was reported to be significantly increased for operation at flux around the critical flux value of the system in its weak definition (Le Clech et al., 2003). Jeison and van Lier (2006), reported that in the long-term operation of a thermophilic AnMBR close to maximum flux, the cake layer is consolidated and backwash is not enough for its removal so additional maintenance is required.

Trans-membrane pressure and flux

Beaubien et al. (1996) explained the relation of the membrane flux to transmembrane pressure (TMP) based on the resistance in-series model. According to it, two main resistances are identified: membrane-solute interaction (R'_m) and concentration-polarization (R_g). The former one includes hydraulic resistance, pore plugging and adsorption; aspects assumed to be very little influenced by operational membrane parameters. R_g includes the effect of gel layer formation and therefore is defined as linearly dependent on TMP. The following equation shows the expected relation of membrane flux and TMP. Mass transfer properties of the system are involved in the factor β of the equation.

$$Flux = \frac{TMP}{\mu(R'_m + \beta\, TMP)}$$

Based on the previous equation, two different operational zones can be identified. When gel layer has little influence in the system, TMP and Flux would be linearly related ($R'_m \gg \beta\, TMP$) while there is a level of gel layer relevance that makes flux independent of TMP ($\beta\, TMP \gg R'_m$). In the experiments of Beaubien et al. (1996) both areas were studied. It was verified that for the low pressure zone, suspended solids concentration and applied TMP are the determinant factors of membrane flux whereas for the high pressure zone a limiting flux was achieved. Additionally, a linear relation between the maximum flux and the cross-flow velocity applied was found.

Considering the aforementioned model, an optimum TMP is found when R'_m and R_g are equal implying that $TMP_{opt}=R'_m/\beta$ which actually is a relation of membrane permeability conditions ($1/R'_m$) and the maximum flux of the system (1/β). Operating at that TMP would lead to the maximum obtainable flux at minimum fouling conditions.

Backwash

Backwash cycles are generally applied as a strategy to reduce reversible fouling in membrane bioreactors, and its efficiency was studied by several authors. Yigit et al. (2009) verified that for an aerobic MBR treating domestic, cycles of 10 minutes with either 15s or 5s of backwash were optimum in terms of fouling prevention and permeate recovery. However, such effect is only observed if operational flux remains below critical values. Additionally, in that study, it was found that backwash frequency has a higher impact on fouling prevention than backwash duration. On the contrary, according to Wu et al. (2008) backwash flux is the prevailing parameter for deterrence of cake layer formation.

2.4.6 Applications in the industry

A classification of wastewaters regarding organic concentration and solubility of contaminants was made by Liao et al. (2006) in order to assess the applicability of AnMBR technology in each case (Figure 2-7). According to the author, the major field of application is for high strength and high suspended solids wastewater as on this substrate the performance of granular and biofilm anaerobic reactors tends to be poor. Retention of particulates in these reactors is not accomplished so degradation of complex particulate COD is not achieved.

On the other hand, conventional high rate anaerobic reactors, especially UASB and EGSB, remain as a more feasible alternative for high strength and low SS wastewater treatment. AnMBRs compared to those involve higher costs of investment (membranes), and operation (pumping), as well as limitation in the OLR of these systems due to their pressure-driven operation. Therefore, only in case very high standards of discharge are imposed, applying membranes for further purification in combination with other high rate systems can be of interest for these wastewaters, (Liao et al., 2006). Additionally, in the event of the presence of toxics or low temperature conditions that might inhibit methanogens, membrane introduction can be considered (Stuckey, 2010).

The low strength wastewaters are mainly related to sewage which is currently aerobically treated with the exception of some warm climate. This is mainly related to the low growth of anaerobic microbial communities at low temperatures. However, high retention of biomass in AnMBR might solve this issue, and thus, there is a potential application of this technology for this kind of wastewater. In comparison to aerobic treatment, despite that not much biogas production is expected in low strength wastewater, savings on aeration energy are considerable, making this technology attractive (Liao et al., 2006).

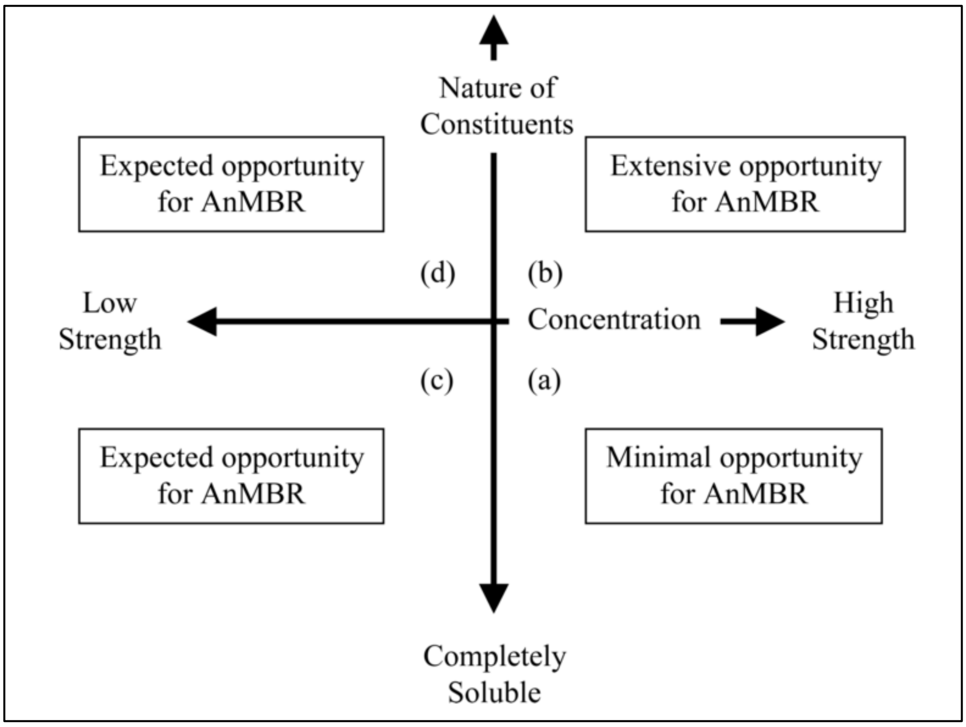


Figure 2-7 Applicability of AnMBR according to wastewater characteristics. Source: Liao et al. (2006)

Considering the previous analysis, the main groups of wastewaters treated in AnMBR in the past decades have been:

- food processing industries like dairy crops processing and beverage;
- industrial (non-food) wastewaters, especially those containing toxic and recalcitrant compounds as pulp and paper, chemical, petroleum and pharmaceuticals;
- high solids streams as sludge from WWT, manure and slaughterhouse wastewaters; and
- municipal wastewater.

Mainly lab-scale experiments have been performed in the last decades operating in a wide range of OLR (up to 24 kg COD/m^3d) and flux (up to 140 $Lm^{-2}h$ (Lmh)). However, extrapolation of results to pilot and full scale might not be correct, especially where hydraulic behaviour and shears are concerned. For instance, so far, although similar fluxes of operation are used, no significant fouling issues comparable to lab scale conditions have been reported for full-scale reactors. Additionally, most full-scale installations have submerged configurations while in laboratory scale external cross-flow membranes are the most common ones (Dereli et al., 2012).

Treatment of cheese whey in AnMBR has been reported. A two-stage reactor consisting of a pre-acidification tank followed by a methanogenic tank coupled with a micro-filtration (MF) membrane was operated by Saddoud et al. (2007). High COD and TSS removals were achieved in the overall system, 98 and 100% respectively. No membrane fouling was reported for the operation. However, the period of study was only of 40 days, so further analysis of long-term operation is required to make conclusions on this issue. Also, other examples of pilot studies treating sweet and acidified cheese whey with similar removal efficiencies were included in the review of Liao et al. (2006).

Regarding dairy industry effluent, in particular, little reports on the application at laboratory, pilot or full-scale are available. Synthetic effluent was treated by Al-Malack and Aldana (2016) in an AnMBR with submerged configuration reaching a maximum COD removal efficiency of 91.4%. However, a very low flux of 2.2 Lmh was maintained in the system to avoid fouling. No studies of the long-term performance of AnMBR treating real dairy wastewater considering biological and filtration performance have been published. Therefore, given the interest in the application of this technology in this industrial sector due to the aforementioned benefits, more research needs to be done in this field.

CHAPTER 3

Materials & Methods

3.1 Reactor set-up

A bench-scale AnMBR in side-stream configuration (Figure 3-1) was run for 107 days. The process tank consisted of a continuous stirred tank reactor (CSRT) with a volume of 10 L. Temperature was controlled by a water bath connected to a water jacket. Continuous monitoring and automatic adjustments of pH were performed by addition of hydrochloric acid (HCl) or sodium hydroxide (NaOH) commanded by a Hach sc100^{TM} controller. Biogas production in the system was measured by a Ritter® wet tip biogas meter. The reactor feeding was done through a peristaltic pump Watson-Marlow 120U controlled by a PLC system. The feed tank was a stirred vessel to ensure particulate matter suspension and homogeneity of the influent. It was kept at ambient temperature to promote pre-acidification of the wastewater (residence time about 3-4 days).

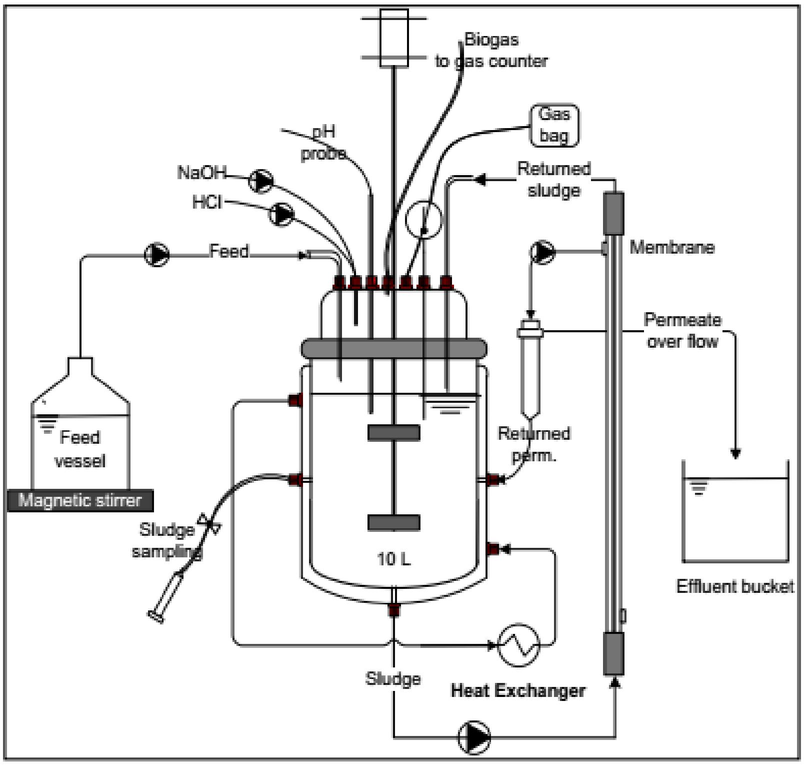


Figure 3-1 Scheme of AnMBR set-up. Source: Veolia-Biothane

Recirculation of the reactor content over a cross-flow UF membrane using a progressive cavity pump (eccentric screw pump) was done in order to separate the biomass and the effluent by decoupling the SRT and HRT of the system. Design characteristics of the membrane used and regular ranges of operation are described in Table 3-1. The cross-flow velocity of recirculation was maintained at 1 m/s during almost the whole operational period. As an exception different values were used during specific experiments are described in section 3.4. A peristaltic pump Watson-Marlow 520 U with adjustable speed was used to pull permeate from the column at a settable constant flux. Permeate recirculation from the collection tank was used to maintain a constant level in the reactor independently of variations in membrane's flux and feeding flows. Effluent from the system was collected in a bucket, and the residual sludge (retentate) from the membrane returned to the reactor.

In order to control fouling due to cake layer formation, sequential backwash was performed by automatically inverting the suction pump's flow direction at defined intervals. Different sequences backwash/filtration, also control by the PLC, were used during the operation but mainly 10s/890s and 15s/900s were the prevalent ones. As an additional measure to help membrane cleaning, the direction of flow inside the membrane was occasionally manually inverted. Clean in place (CIP) of the membrane was done on demand, after evaluation of the performance of the system.

Table 3-1 Membrane characteristics. Source: Veolia-Biothane

Type of membrane	**Ultrafiltration**
Geometry	Tubular
Flow type	Cross-flow
Filtration mode	Inside to outside
Manufacturer/Model	Pentair Xflow
Material	Polyvinylidene fluoride (PVDF)
Hydrophobicity	Hydrophobic
Effective surface area	0.049 m^2
Diameter	5.2 mm
Mean pore size	30 nm
Design flux	30 Lmh or higher
Pressure/suction range	0.5 – 2 bar

3.2 Methodology

3.2.1 Substrate characterization

Process effluent and cheese whey effluent from a dairy processing industry were used for reactor's feeding. During the whole operation, 2 batches in total were received and preserved in cold storage room at 5°C. Both of them were characterized at arrival to the laboratory. Characterization included the following parameters: TCOD, SCOD, total suspended solids (TSS), volatile suspended solids (VSS), pH, VFAs, total Kjeldahl Nitrogen (TKN), soluble Kjeldahl nitrogen (SKN), ammonium (NH_4^+), anions (Cl^-, NO_3^-, NO_2^-, SO_4^{2-} and PO_4^{3-}), cations (Na^+, K^+, Mg^{2+} and Ca^{2+}) and total phosphorus (P).

3.2.2 Feeding preparation and regime

A mixture of process effluent and cheese whey in a relation of 4.5:1 was used in order to simulate the relation of flows generated by the dairy industry of study. Two extra solutions were added in order to ensure the presence of all nutrients required for growth:

- $FeCl_3$ + Vithane®; 0.105 g solution/gCOD
- Ca/Mg/K; 2 mgCa/gCOD, 3 mgK/gCOD & 4 mgK/gCOD

However, the addition of Ca/Mg/K was interrupted after the first 3 weeks as it was verified that the amount of those elements present already in the raw wastewater were sufficient for growth.

A feast-famine regime consisting of 1 hour of feeding followed by 5 hours of reaction was applied. The VLR was maintained at an average of 6 ± 1 gCOD/(L·d) during the whole period of study by changing the daily feed flow between 3.1 to 4 L/d according to the feed concentration obtained in every preparation.

3.2.3 Operational conditions

Selected operational conditions for the system are summarized in Table 3-2.

Table 3-2 Operational parameters

Parameter	Value
Temperature (T)	35 ºC
pH	6.5 – 7.5
Permeate flux	Variable (according to set point)
SRT	30 days
VLR	6 g COD/(Ld)

3.2.4 Reactor history & “start-up”

At the time this research was started, the reactor had been operating for 425 days fed with synthetic wastewater based on whole milk. The reactor was originally inoculated with crushed and sieved granular sludge taken from food processing companies (PURAC and MARS). The following two phases of operation were applied for the synthetic feeding period (not part of this MSc research):

- Phase I (0 to 245 days): continuous feeding at a VLR of 4-5 g COD/Lday and SRT of 40 days.
- Phase II (245 to 425 days): sequential feeding (1 hour feed: 5 hours reaction) at two different operational conditions. First, VLR of 5 g COD/Ld and 40 days SRT (day 245 to 320) and second, VLR of 6 g COD/Ld and 30 days SRT.

In order to avoid disturbances in reactor operation due to the substrate change, a VLR of 4 g COD/Ld was applied during the first week with the real dairy wastewater. Given that the VFA level remained at very low values, the VLR was increased to 5 g COD/Ld after 6 days and to 6 g COD/Ld on the 7^{th} day and remained in that condition for the rest of the operation.

Table 3-3 Performance of the AnMBR fed with synthetic dairy WW before changing to real WW. Source: Veolia-Biothane

Feeding		Operational conditions		Operation results	
Flow (L/d)	6	HRT (h)	40	COD removal (%)	99.4
TCOD (g/L)	10.5	SRT (d)	30	Biogas prod. (NL/d)	17
SCOD (g/L)	2.8	VLR (gCOD/Ld)	6.0	CH_4 content (%)	72
FOG as COD (g/L)	4.9	TS sludge(g/L)	12.3	CH_4 (gCOD/Ld)	5.8 (55%)
TS (g/L)	4.2	VS sludge (g/L)	10.8	TMP (mbar)	500
VS (g/L)	3.7	Flux set point (Lmh)	17	Real flux (Lmh)	11

3.2.5 Follow-up, sampling and analysis

Samples were taken at regular basis from the reactor, the feed tank and permeate line or vessel to evaluate the system's performance. The following Tables 3-4 and 3-5 summarize parameters of operation registered from the software and analysis performed (including frequency) for the follow-up op the biological performance and sludge filterability respectively.

Table 3-4 Biological performance follow-up parameters

Parameter	Unit	Sample and frequency				
		Feed (old)	Feed (fresh)	Sludge	Permeate	Biogas
pH	-	w	w	5/w		
T	^0C			c		
Biogas volume	L					c
Biogas CH_4 content	%					5/w
Total COD	gCOD/L	w	w	w	2/w	
Soluble COD	gCOD/L	w	w	w		
TS & VS	gS/L	w	w	w	2/w	
TSS & VSS	gSS/L	w	w	w		
VFA	mgVFA/L	2/m	2/m	w	2/w	
TKN	mgTN/L	2/m	2/m	2/m	2/m	
SKN	mgSN/L	2/m		2/m		
NH_4^+	mgN/L	2/m	2/m	2/m	2/m	
Anions	mg/L or mmol/L	m		m	m	
Cations	mg/L or mmol/L	m		m	m	

Parameter	Unit	Sample and frequency				
		Feed (old)	Feed (fresh)	Sludge	Permeate	Biogas
PO_4^{3+}	mgP/L	2/m		2/m	2/m	
P Total	mgP/L	2/m		2/m	2/m	
SMA	$gCOD_{CH4}$/(g VSS d)			2/m		
Alkalinity	meq/L			w	w	

Reference: c-continuous, x/w-x times a week, w- weekly, x/m- x times per month, m-monthly

Table 3-5 Sludge filterability characteristics follow-up tests

Analysis	Unit	Sample: Sludge Frequency
Capillary suction time (CST)	S	2/w
Viscosity	Pa·s	1/w
Relative Hydrophobicity*	%	2/m
Morphology*	-	2/m
PSD	-	Variable (5 times)

Reference: x/w-x times a week, x/m- x times per month/*Analysis to be performed at KU Leuven according to collaboration

Furthermore, in order to evaluate membrane permeability, transmembrane pressure (TMP) was continuously registered, and the flux was measured 5 times per week.

3.2.6 Evaluation of permeability and flux for different cross-flow velocities and backwash frequencies

Specific experiments were performed during last 5 weeks of reactor operation in order to evaluate the effect of increasing either cross-flow velocities or backwash frequencies on the flux and permeability of the system.

Experimental conditions

The specific methodology for the experiments was established as follows.

- Stable biological operation at a constant VLR of 6 gCOD/Ld.
- Sludge properties, namely MLSS, CST, viscosity and PSD, were evaluated once or twice a week during the trials.
- Considering the intermittent feeding regime, the total volume of the reactor varies and consequently MLSS concentrations (see section 4.2.8). To avoid that dilution effect, the trials were run during the period of constant volume in the reactor, so one hour after finishing the feeding phase.
- Similar TMP was used for all the different conditions. This was achieved by two main actions: manual pressure regulation of the recirculation line by a valve installed in the retentate side (to values of highest cross-flow velocity) and constant pressure in

permeate line by opening the permeate pump during filtration. Pressure data was obtained from the files generated by the PLC of the system (Biothane AnMBR Application).

- Backwash was done at a constant duration (15s) and flux (200 Lmh) through the permeate pump. The pump was manually closed every time at the end of the cycle and re-opened again to start the following cycle. Backwash flux was verified every day before starting the trials (see Appendix B for further details).
- Regarding backwash frequency, for every trial, the system was run at the selected condition at least from the day before the experiment. This was done trying to reach certain steady state (short-term) on membrane performance for the different cycles.
- Cross-flow velocity was experimentally verified before starting every trial. Measurements were started after at least 3 cycles of operation with the selected cross-flow velocity to ensure stable conditions.
- The membrane flux was continuously measured in real-time during 3 filtration cycles using a Kern Scale connected through Kern Balance Connection Software (see Appendix B for further details).
- Tests of clean water permeability (CWP) were performed daily on the membrane using a setpoint of 50 Lmh and a cross-flow velocity of 1 m/s on the system. This was done in order to assess membrane conditions during different factors conditions (see Appendix B for further details).
- All factor's levels were tested in triplicates. In case of relevant variations on sludge characteristics or membrane permeability, tests were repeated to mitigate the influence of those parameters on results.

Experimental design

Considering two different cycle durations and four levels of cross-flow velocity were selected for evaluation, a total of 8 different experimental conditions were derived. According to the limitations defined in the methodology presented in the previous section, one condition was tested per day using a random order for the first set of experiments. According to results of membrane clean water permeability and sludge characteristics, some operational conditions were re-tested to get more comparable results. The logic behind these decisions will be further explained in section 4.5.2 where results are presented. The schedule of the different trials is included in Appendix B.

Two-way analysis of variance (ANOVA) was used to evaluate the significance of the effects of every individual factor as well as the existence of interaction among factors. Additionally, the conditions of the membrane and the sludge during trials were compared for every set using Grubbs test for identification of outliers at a significance level of 95%.

3.3 Analytical methods

The analyses defined in previous section 3.2.5 was performed in accordance with Biothane® existing analytical procedures, see also Table 3.6.

3.3.1 Online monitored parameters

The following parameters were continuously registered by the PLC:

- pH,
- temperature,
- biogas production, and
- filtration pressures (TMP, feed pressure, retentate pressure and permeate pressure).

3.3.2 Physicochemical analysis

The analytical methods applied for the determination of the rest of the physicochemical parameters of the samples are summarized in Table 3-6.

Table 3-6 Analytical methods of physicochemical analysis

Parameter	**Analytical method**
Total COD	HACH Lange: 514,914 or 014 (to be defined according to sample concentration)
Soluble COD	HACH Lange: 514,914 or 014 after filtration with glass fibre filter
TS	Standard method gravimetric analysis
TSS & VSS	Standard method gravimetric analysis after filtration with glass fibre filter
VFA	Quantitative gas chromatography
TKN	Chemical decomposition, distillation and titration
SKN	Chemical decomposition, distillation and titration after filtration with glass fibre filter
NH_4^+	Distillation and titration
Anions	External laboratory
Cations	External laboratory
PO_4^{3+}	HACH Lange 049
P Total	HACH Lange 350
Biogas CH_4 content	Portable biogas analyser (weekly calibration)

3.3.3 SMA

This test was performed using Oxitop® equipment. Sludge was mixed in test bottles (duplicate) with sufficient amount of easily biodegradable substrate (acetate). Amount of sludge and acetate used were standard according to Biothane® procedures for all analysis (49.4 g of sludge and 0.6 g acetate solution).

Bottles were flushed with a mix of N_2:CO_2 (70%:30%) in order to establish anaerobic conditions during the analyses. A rotary shaker located in a temperature controlled dark chamber (35°C) was used for incubation. Either 2 or 3 pulses of acetate were used for the calculation of the average activity in each analysis. Methane production vs time was calculated from Oxitop®'s heads pressure data assuming 0.35L of methane production per gram of COD and total adsorption of CO_2 in the heads. The final activity value was calculated using the slope during the linear phase of the methane production and the results of VSS analysis.

3.3.4 Sludge filterability tests

Capillary suction time

The capillary suction time is a measurement that indicates the dewaterability properties of the sludge considering the time it takes for the filtrate to permeate through a paper filter. It is measured by the time it takes for water from the sludge sample to travel from one electrode to another in a standard cell. The equipment used was a Triton Capillary Suction timer with a special filter paper from Triton Electronics Company. All measurements were performed at least by duplicate with the sludge at 37ºC.

Viscosity

This parameter is a measure of the resistance of the sludge to flow which also contributes to characterize its filterability. The equipment used was HAAKE Viscotester® 550 which is capable of measuring viscosity at different temperatures being equipped with a water bath for temperature control. All the measurements were done at 37ºC.

Particle size distribution (PSD) analysis

A Bluewave laser diffraction analyser from Microtac was used for the determination of PSD in the sludge from the reactor at different stages of operation. The measurement range is from 0.01 to 2800 µm and the equipment counts with a special algorithm that allows computation of irregular shapes and also transparency and absorption properties of particles (Microtac, 2018).

One measurement was run after the first month of operation and 5 different measurements (one per week) were done in the last month of operation during specific experiments performed in the reactor. The selected flow in the equipment for the analysis was of 25% of maximum as it was verified that for higher flows the particles were damaged (see Appendix A). The regular protocol for sludge evaluation performs 3 runs for every sample taken and reports the average values. Samples were analysed by triplicate in all cases, and the average values of the 3 runs computed as the final result.

Morphology evaluation (by KU Leuven CREaS)

Several microscope images from sludge diluted to 1 g MLSS/L were taken with an inverted Olympus IX83 microscope. Software developed by KU Leuven CREaS division, Activates Sludge Image Analysis Program (ASIAP) was used for image analysis. The output of the image evaluation includes particle size distribution, total filament length, the relative number of flocs, relative floc area and floc shape parameters.

Relative hydrophobicity (by KU Leuven CREaS)

For the determination of the relative hydrophobicity of the sludge, extraction with hexane was used based on the affinity principle. After extraction of sludge from the water phase, transparency/turbidity was measured in a UV-VIS spectrophotometer using 650nm wavelength. The higher the transparency of the water phase after extraction in comparison to the original sludge, the higher the relative hydrophobicity of the sample.

Scanning Electron microscopy (SEM) & Energy-dispersive X-ray Spectrometry (EDS)

Sludge and Crystals obtained from the reactor were analysed using a SEM JEOL JSM 6610A combined with EDS in 3 different dates. SEM allows clear observation of surfaces that cannot be distinguished by an optical microscope. The main principle is the use of electrons that have a shorter wavelength than light, allowing resolutions up to 0.5 to 4 nm versus 200 nm in the regular microscope. Additionally, it enables 3-D image observation as it has a deeper focal depth. Furthermore, it was combined with EDS for elemental analysis of surfaces in order to determine especially the crystals composition.

CHAPTER 4

Results and discussion

Results of reactor operation are presented and critically reviewed in this chapter. A total of six different sections were defined including substrate characterization, biological performance, sludge characteristics, membrane performance, optimization of membrane operation and overall discussion. The first four sections include analysis of the different aspects of reactor performance during the 107 days of operation with dairy wastewater. The fifth section presents the results of the evaluation of cross-flow velocity and filtration cycles on the obtained flux at constant TMP for this system. Some considerations for improvement of the existent set-up are also presented in that section. A review and discussion of the main results from this thesis are included as the last section in this chapter.

4.1 Substrate characterization

4.1.1 Main streams composition

During the period of reactor operation, 2 different batches of wastewater (wash water) and whey from a cheese-producing industry in Greece were received. The average values of characterization at arrival are included in Table 4-1. Some variations on compositions were expected among the different barrels of WW and also some degradation even with storage at 5ºC. Therefore, results of regular analysis were done on the actual feed prepared by mixing 4.5 parts of wash water and 1 part of whey and values are presented in the same table.

Both cheese whey and wash water COD and SS values measured are within the range of reference values shown in the reviews of Carvalho et al. (2013) and Demirel et al. (2005). The COD fractionation of the whey presented high variability from one batch to the other, ranging from 60 to 95% of soluble COD in the stream. This difference could be related either to the wastewater collection procedure or the production of different classes of cheese. On the other hand, wash water from both batches presented similar characteristics which contributed to having more stable values in the prepared feed. However, full-scale reactors' operation might require closer attention to high variations of these streams to avoid affecting the biological performance of the treatment system.

In terms of nutrients availability, for the mixture to be treated, the relation COD:N:P was 200:14:3. This amount of N and P covers the requirements for biomass growth that range from 400:7:1 up to 1000:7:1 depending on the sludge loading rate and feed VFAs content of the system (Henze and Harremoës, 1983; Kolev Slavov, 2017). Regarding micronutrients, as expectable from a food processing residue, their quantity is sufficient to allow microbial growth. However, divalent cations concentrations are lower than the advisable values for good flocculation of 100-200 mg/L and 75 – 150 mg/L of Ca^{2+} and Mg^{2+}, respectively (Kugelman and McCarty, 1965).

Table 4-1 Characterization of wastewater streams at arrival

Parameter	Whey	Wash water	Prepared Feed
pH	4.3	4.1	3.7 - 4.3
COD (g/L)			
TCOD	69 -104	3.6 - 4.1	15.6 - 20.0
SCOD	59- 66	2.8 - 2.9	12.9 - 17.7
PCOD	45	0.7 - 1.2	1.2 - 6.0
Solids (g/L)			
TS	58- 62	4.1 - 4.7	-
VS	54 - 56	2.6 - 2.9	-
TSS	1.5 - 7.8	0.29 - 0.30	0.8 - 2.6
VSS	1.4 - 7.6	0.28 -0.29	0.8 - 2.2
Nitrogen (mgN/L)			
TKN	1800 - 1900	114 - 165	436 - 479
SKN	1760 - 1850	70 - 147	360
NH4-N	120	78	20 - 40
Phosphorous (mgP/L)			
P-tot	402 - 436	60 - 113	20 - 173
ortho-P	216 - 312	51 - 99	36 - 120
Volatile Fatty Acids (meq/L)			
C2 : C6	3.1	1.5	4 - 9
Major Anions (mg/L)			
SO42-, sulphate	538 - 643	58 - 67	115 - 154
NO3-, nitrate	12	43 - 136	31 - 112
NO2-, nitrite	< 0.1	0.1 - 10	<0.1
Cl-, chloride	909 - 912	316 - 579	472 - 589
Major Cations (mg/L)			
Na+, sodium	334 - 421	258 - 380	267 - 384
Mg2+, magnesium	100 - 117	24 - 27	34 - 41
K+, potassium	1419 - 1670	78 - 129	254 - 348
Ca2+, calcium	341 - 441	112	144 - 164
Others			
Conductivity (mS/cm)	5.54	2.55	-
TOC (mg/L)	25600	1251	-
Alkalinity Total (meq/L)	13	3	0 - 3
Alkalinity HCO3 (meq/L)	0	0	0
Density (g/mL)	1.0216	1.0005	-
FOG (mg/L)	409 - 1124	400 - 700	530 - 650

The pH of the wastewater is an important parameter to consider as it can inhibit the anaerobic digestion process, especially methanogenesis which is optimum at a pH range of 6.8 – 7.2 (Nadais et al., 2010). In this case, both wash water and whey presented low pH values at arrival compared to on-site measurements obtained were 7.9 and 6.1, respectively. This difference is likely related to pre-acidification which occurred during transport from the production facility in Greece to the Netherlands. The low levels of alkalinity generally present in dairy wastewater (Kolev Slavov, 2017) entail a rapid decrease in pH under fermentative conditions.

The results of VFAs concentration measured with the quantitative gas chromatography method did not show high acidification. However, only C2: C6 acids (formic, acetic, propionic, isobutyric, n-butyric, isovaleric, n-valeric, isocaproic and n-caproic) were detected through this method. Considering the high amount of lactose in whey (~70% of dry weight; Kolev Slavov (2017)) and wastewater (~30% of COD; Fang and Yu (2000)) lactic acid, which was not quantified, is expected to be the main product of acidification. Total VFAs measurements by titration can be performed to verify this idea. It is advisable to do so before up-scaling the system as pH is a crucial characteristic of anaerobic systems. The other possibility is that the facility produces WW with very different pH and thus, homogenization before treatment would be necessary.

As shown in Table 4-1, almost no alkalinity is available in the received wastewaters and this can lead to unstable performance of the anaerobic process. However, it should be considered that the degradation of organic nitrogen compounds in the system is an additional source of alkalinity (Ekama, 2017). Given the organic nitrogen content of this feed, and assuming its total release as ammonia, considering 1 meq-Alk/14gN, there is a potential for alkalinity generation of 28 meq/L, which corresponds to 1400 mg Alk-$CaCO_3$/L. The pH in the reactor is a result of the alkalinity and the partial pressure of CO_2. Analysing Figure 4-1 from McCarty (1964), those levels of alkalinity allow operation of the reactor within normal levels at a pH range from 6.6 to 7, depending on CO_2 concentration. Since this reactor operates in sequential feeding regime, an increase in VFAs is expected during feeding phase and with this alkalinity availability on the low limit of acceptable operation, pH control might be necessary.

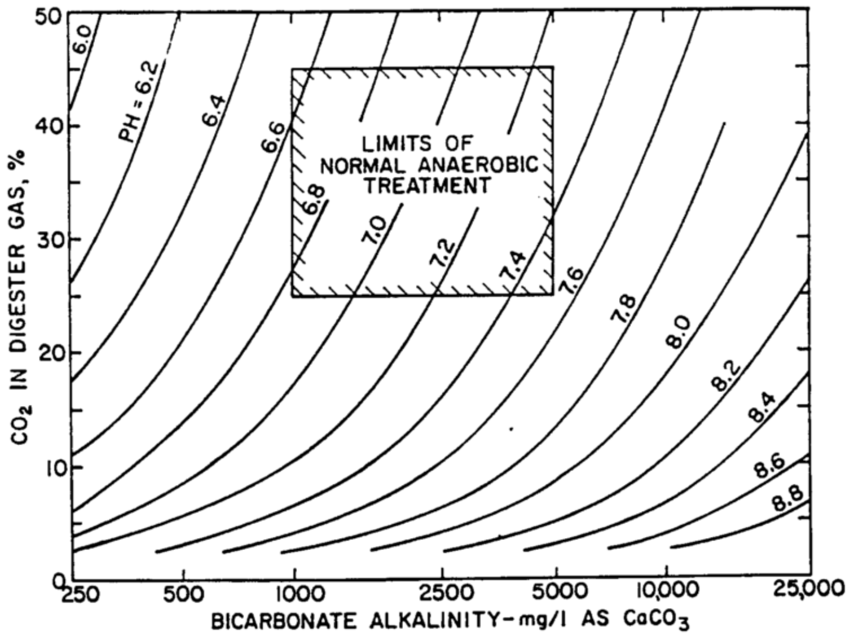


Figure 4-1 Reactor pH vs bicarbonate and CO_2 concentration at 35ºC. Source: McCarty (1964)

Regarding FOG content, the values for this wastewater are lower than the ones measured for the synthetic substrate used during the previous stage of reactor operation (see Table 3-3). While for the synthetic it corresponded to 4.9 g FOG-COD/L (1.8 g FOG/L), representing almost 50% of total COD, in the new wastewater, it is ~1.5 g FOG-COD/L (0.6 g FOG/L) representing only 10% of the total COD. However, this FOG concentration is still considered as high for treatment in granular systems without a pre-treatment step (Leal et al., 2006; Passeggi et al., 2012). For that reason, benefits of AnMBR over granular systems still come into effect for this effluent.

4.1.2 Feed pre-acidification

Pre-acidification is commonly applied to improve the efficiency of anaerobic treatment for industrial wastewaters. Two-phase reactors with controlled conditions to optimize acidification have been successfully applied both for cheese whey and dairy wastewater accomplishing higher methane production and COD removal (Demirel et al., 2005; Karadag et al., 2015b; Yang et al., 2003).

In this case, the feed was prepared twice a week keeping always approximately one-quarter of old feed that acted as an inoculum. This, together with the observed degradation of the substrate even in the fridge (gas formation) explain the difference in VFA values measured on prepared feed compared to the streams at arrival. The feed bucket was continuously mixed and kept at room temperature for an average of 3-4 days, and even though conditions were not optimized for pre-acidification, it is still expected to take place to some extent.

Results of the analysis of pH, COD and VFAs on freshly prepared feed and old feed performed weekly are shown in Table 4-2. A total increase from 3% to 11% of VFAs in relation to COD content was observed. Acetic and propionic acid accounted for almost 100% in both fresh and old feed. Additionally, the slight decrease in the pH and COD observed proved that there is certain degradation in the feed tank over the days. However, the values were low considering the long residence time compared to two-phase reactors were the HRT of the acidogenic phase is generally between 12 and 24 hours (Demirel et al., 2005).

For acidogenesis of lactose, which is the main constituent of whey and dairy wastewater, pH was reported as the key leading factor by Kisaalita et al. (1987). The study showed that the pH should be maintained above 4.5 to avoid the formation of CO_2, H_2 and ethanol instead of acetate. The maximum formation rate of VFAs, and therefore optimum fermentation condition, was found at pH values of 6 – 6.5. In this case, no pH control was performed in the feed bucket, and consequently, the low pH of the feed was identified as the main factor limiting the acidification into acetate. Additionally, as mentioned in section 4.1.1, pre-acidification is suspected to have occurred during transport of the wastewater from the production facility to the laboratory.

Table 4-2 Characteristics of fresh and old feed

Characteristic	Fresh feed	Old feed
TCOD (mg/L)	15.6 - 20.0	11.0 – 18.5
SCOD (mg/L)	12.9 - 17.7	9.9 – 16.6
pH	3.7 – 4.3	3.4 – 4.1
VFA (meq/L)	4 – 9	13.8 – 28.7
VFA (%COD)	~3%	~11%

4.2 Biological reactor performance

4.2.1 VLR, SLR & SMA

The operational VLR for the reactor during the whole period of study was set at 6 g COD/Ld. Regular variations on COD concentrations of the feed resulted in a real value of

(5.8 ± 1.5) g COD/Ld. As mentioned in section 3.2.4, during the first week of operation with the real wastewater, the VLR was kept around 4-5 g COD/Ld as it can be noticed from Figure 4-2. Additionally, between days 45-52 the company remained closed and a failure on recirculation pump, due to solids accumulation and clogging of the suction, forced to stop the reactor. After that period the reactor was cleaned to remove the accumulated precipitants and operation was re-established to normal VLR on day 65.

The applied VLR is within the range of existent applications of anaerobic technologies reported for treatment of dairy wastewater with similar characteristics. Gavala et al. (1999) treated wastewater from a cheese industry in a lab-scale UASB reactor finding 6.2 g COD/Ld as the most stable value, reaching up to 7.5 g COD/Ld. A much higher loading rate of 28.5 g COD/Ld was applied on the treatment of raw cheese whey of 60 g COD/L in another lab-scale UASB reactor. However, during the first phase when cheese whey wastewater with a concentration of 14 g COD/L was used, the VLR was kept at a maximum of 2.5 g COD/Ld (Kalyuzhnyi et al., 1997) due to further limitations in the HRT.

Two-phase anaerobic reactors are considered in general to be more efficient for the treatment of dairy effluents reaching higher loading rates at lower HRTs. The presence of lactose in this effluents makes the acidification phase short (in general around 24h) and efficient, producing VFAs that can be easily degraded in the methanogenic stage (Britz et al., 2006). Particularly, an AnMBR with a pre-acidification reactor was used by Saddoud et al. (2007) for the treatment of acid cheese whey (68.6 g COD/L) reaching total VLRs up to 20 g COD/Ld.

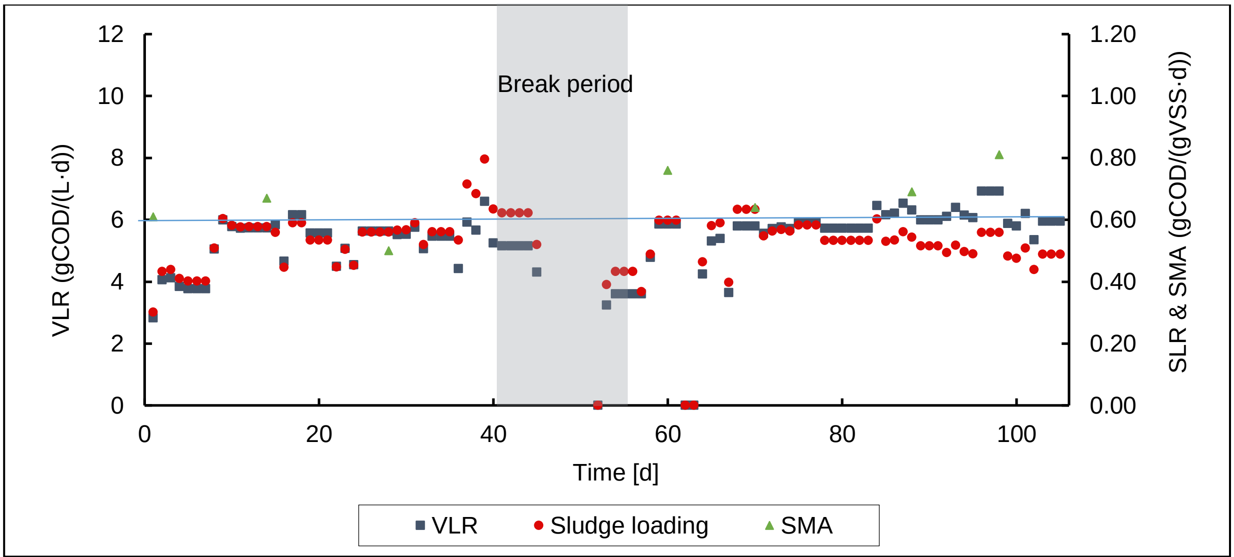


Figure 4-2 Volumetric loading rate, sludge loading rate and specific methanogenic activity

An additional operational parameter that is not commonly reported, but defines the performance, is the sludge loading rate (SLR). In the case of high strength wastewaters, for which the HRT is not generally limiting the degradation, the SMA of the sludge in the reactor limits the maximum load that can be applied to the available biomass (Ince et al., 1995). For this reactor, the SLR used ranged from 0.5 – 0.6 gCOD/gVSSd while the SMA measured on acetate was between 0.6 - 0.8 gCOD/gVSSd (Figure 4-2). This means that probably higher SLR can be applied in this reactor as enough active biomass is available for degradation.

Reported values found for suspended sludge in other AnMBR are lower than the ones measured in this study. Ince et al. (1995) reached 0.14 gCOD/gVSSd (at 36°C) for brewery WW, while for municipal WW, activities from 0.12 – 0.19 gCOD/gVSSd (at 25°C) were achieved by Ho and Sung (2010). Conversely, the activities measured in the present study are within the range of activity for granular sludge, which is between 0.1 – 2 gCOD/gVSSd (van Lier et al., 2008).

4.2.2 VFAs

Monitoring VFAs concentration in the effluent of anaerobic reactors is generally used as the strategy to control the system performance. This is based on the principle that an overload of the system will be initially shown by VFAs accumulation because they are the substrate of the methanogenic community which is generally the limiting one. In case VFAs concentrations are higher than a defined value, feeding should be stopped until they are consumed, and the organic load should be revised. However, the maximum admissible value for VFAs varies for different characteristics as also the extent of inhibition depends on the type of VFA and sludge (Franke-Whittle et al., 2014). A limit of 5 meq/L was used in this particular case based on the experience of the company with this kind of wastewater treatment.

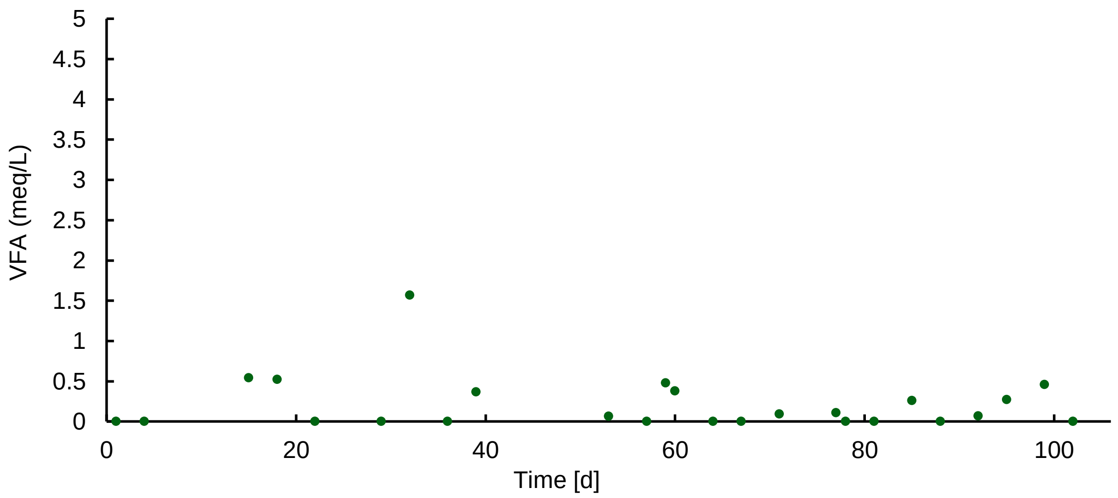


Figure 4-3 VFA concentration in reactor's effluent

As it can clearly be seen in Figure 4-3, the system was never overloaded during operation as the VFA concentrations were almost zero reaching a maximum value of 1.8 meq/L. It is important to mention that variation of different parameters during one complete feeding-fasting cycle was studied showing, as expected, an increase in VFAs during the first hours after feeding. Consequently, in order to assess the real capacity of the system, samples for VFA quantification were always taken from the permeate line close to the end of the fasting period.

The current values of sludge activity and the very low VFAs show that, as far as biological performance is concerned, higher values of VLR could be tested. Considering SMA is maintained, up to a 8 gCOD/Ld could be degraded with the current sludge concentration in the reactor. However, it should also be considered that activity for complex substrates might be lower than SMA and also the filterability characteristics of the sludge can be affected by changing the applied loading rate.

4.2.3 HRT & SRT

The hydraulic retention time was in average 2.7 days and ranged from 2.4 to 3.3 days (Figure 4-4) as the flow was adjusted to compensate for the variations of feed concentration in order to maintain the desired VLR of operation. The HRT reported for previous experiences of anaerobic treatment of dairy WWs of medium to high concentration varies from 20 hours up to 11 days (Demirel et al., 2005; Kolev Slavov, 2017).

Given the capacity of AnMBRs to retain solids, and therefore particulate COD fraction, shorter HRTs are expected to be successfully applied. However, highly active biomass is required to maintain short HRT with proper COD removal efficiency. So far HRTs used in AnMBR are still higher than in other high-rate anaerobic reactors (Liao et al., 2006). An HRT of 4 days was used for the treatment of whey in an AnMBR with a previous acidification phase (Saddoud et al., 2007). Even though two-phase reactors are supposed to require lower HRTs, the effluent used in this experiment was raw whey of a higher concentration so the limit might have been given by SLR and sludge SMA.

In general, applied HRT for anaerobic treatment of whey containing effluents is higher than for other dairy streams, reaching up to 2 days (Prazeres et al., 2012). Gavala et al. (1999) treated wastewater from a similar origin to the one in this research using an UASB reactor at a comparable VLR of 6.2 g COD/Ld achieving 98% of COD removal operating at an HRT of 6 days. Considering that concentration for that experiment was double (37 g COD/L), the WW should have contained a higher proportion of whey.

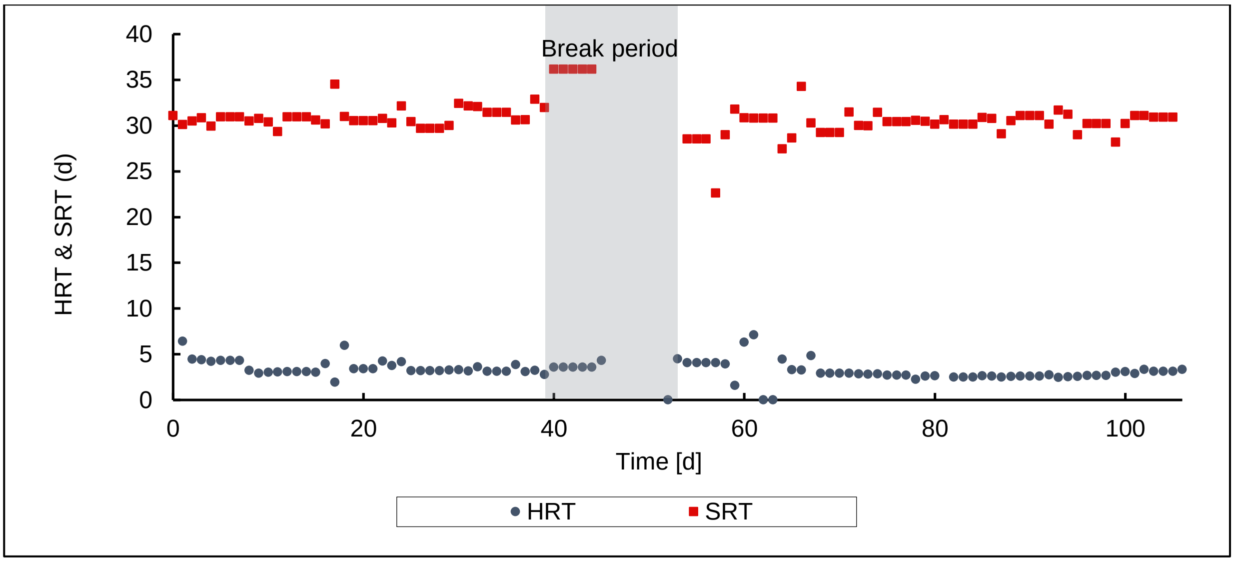


Figure 4-4 Hydraulic and sludge retention time of the system

The SRT of this system was set as 30 days (Figure 4-4) and was controlled by manually wasting sludge from the reactor in order to avoid differences due to variations in biomass concentrations. In contrast to granular sludge systems were high SRTs of 100-200 days can be applied, negative effect of high SRTs was proven for AnMBR (Dereli et al., 2012). Different factors as higher solids, SMP and inorganics concentrations are related to a lower membrane flux and higher fouling (Dereli et al., 2012; Jeison and van Lier, 2006). Huang et al. (2011) found a significant difference in MLSS concentration, as well as an increasing TMP while increasing SRTs from 30 days to 60 days and ∞ time. Additionally, serious fouling was related to SRTs higher than 50 days applied for food processing wastewater treatment (He et al., 2005; Jensen et al., 2015).

4.2.4 Operational pH

Two different phases for the pH of operation in the reactor can be identified from Figure 4-5. Before the failure of the pump during the break due to the accumulation of crystals, an average pH of 7.0 was maintained in the reactor consuming around 1.8 meq/g COD equal to 31.3 meq/L of effluent. Given the identification of high amounts of calcium phosphate precipitates by SEM-EDS analysis, it was decided to drop the pH set point in order to avoid base addition. Operation in those conditions would also be very valuable for full-scale applications as would mean a sharp decrease in the operational costs.

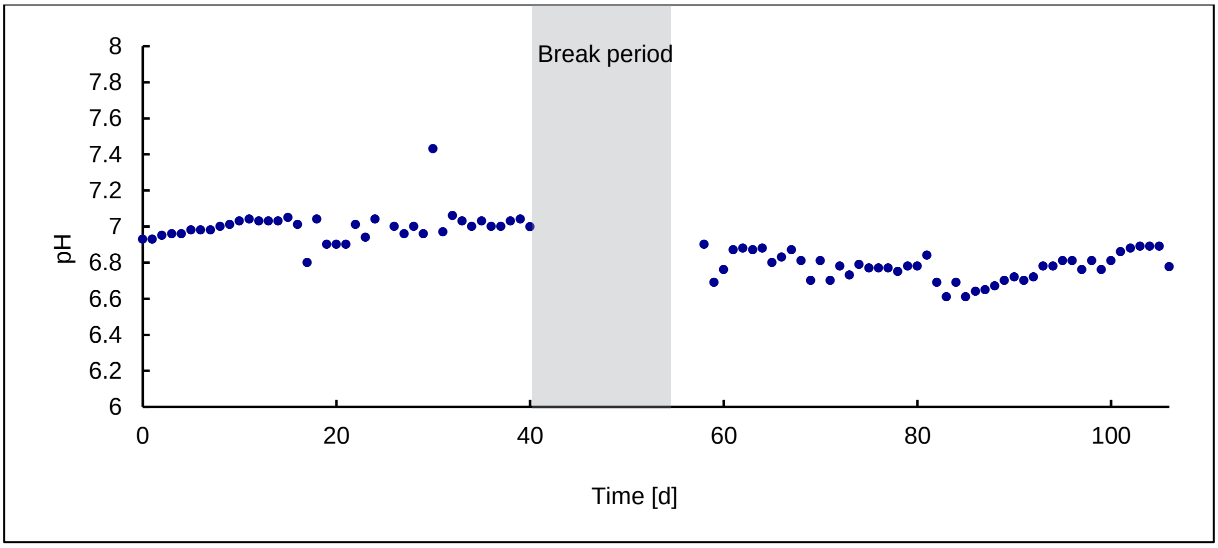


Figure 4-5 Operational pH in the reactor

Due to the aforementioned reasons, the average operational pH after the break was 6.8, reaching a minimum of 6.6. The base addition system was only kept for emergency reasons. No base consumption was registered as the minimum pH to start base addition was set at 6.4 while the minimum value registered in the system was 6.5 at the end of feeding phase (see section 4.2.8). By no addition of a base, the pH in the reactor was determined by the existent alkalinity in the bulk liquid and the partial pressure of CO_2. Results of measured alkalinity in the effluent and CO_2 partial pressure in the headspace are summarized in Table 4-3. The average value of alkalinity matches the estimation from degradation of organic nitrogen presented in section 4.1.1. The pH measured in the reactor was in accordance with the prediction from carbonate equilibria (Figure 4-1).

Table 4-3 Measured alkalinities, CO_2 concentration and estimated pH

Parameter	Average	Minimum	Maximum
Bicarbonate alkalinity (mgCaCO3/L)	1450	1000	2000
CO2 (%)	31.0	25.1	37.4
Estimated pH from equilibrium	**6.8**	**6.65***	**7.1***

*Minimum value calculated from max CO_2 and maximum from minimum CO_2

No negative effects on biological performance of the reactor were observed due to the change in operational pH as no drops on the methane conversion rate or SMA of sludge were detected.

However, deterioration of filterability properties of the sludge might be related to this factor as will be further explained in section 4.3.1.

4.2.5 Solids concentration

The suspended solids concentration in the reactor was mainly constant at average values of TSS and VSS of 11 g/L and 9.5 g/L, respectively during the first 2 months of operation (up to day 70). For that period VSS represented 87% of TSS, and TSS/TS was 84%. For the last month of operation higher values of TSS and VSS were established reaching 12.8 g/L and 11.0 g/L, respectively. The VSS/TSS relation was maintained around 86% while TSS/TS ratio increased to 90%. This increase might be related to a reduction in dissolved solids due to the interruption of base addition.

The TSS measured are similar to previous values reported for dairy wastewater. Saddoud et al. (2007) reported values from 6.4 to 10 gVSS/L with a range of SRT from 30 to 70 days for the treatment of cheese whey in a cross-flow AnMBR. A synthetic effluent made from sucrose and dried cheese whey was treated in a submerged AnMBR by Spagni et al. (2015) at a TSS concentration varying from 8 to 10 g/L with an SRT of 50 days.

Suspended solids concentrations in dairy wastewater treatment seem to be lower when compared to other substrates. A steady-state value of 17.2 gTSS/L was measured in the treatment of wastewater from corn-based ethanol production also at 30 days SRT (Dereli et al., 2015). The same TSS concentration for the steady state was measured in the treatment of palm oil effluent in another AnMBR (Abdurahman et al., 2011).

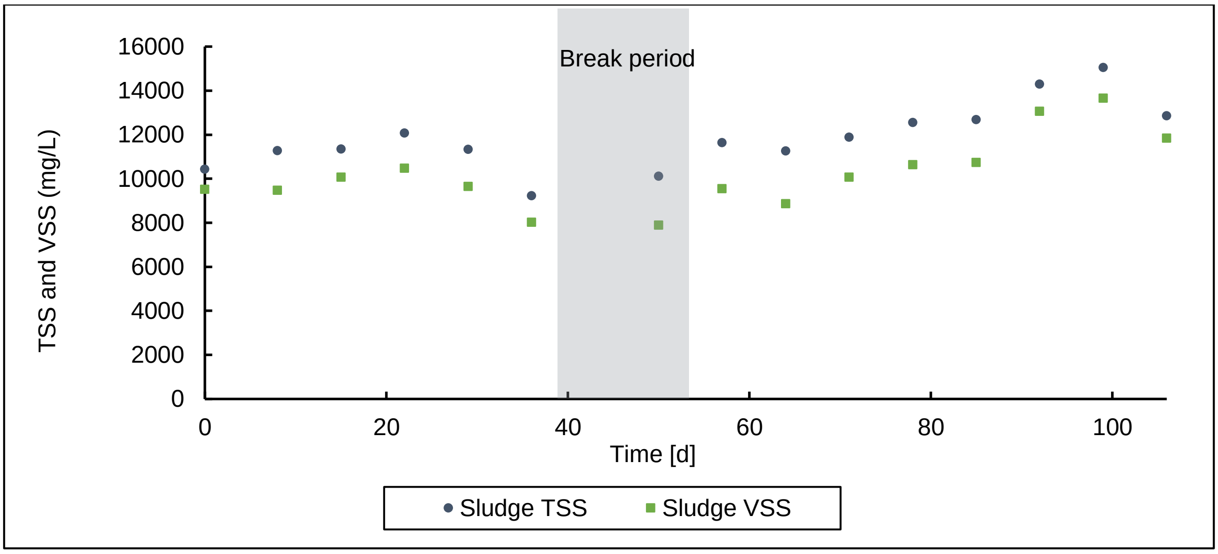


Figure 4-6 Evolution of Total and Volatile suspended solids concentration in the reactor

For the operation of AnMBRs, suspended solids gain importance as the obtained flux was found to be related to TSS of the sludge by many authors. For the operation of submerged membranes, Jeison and van Lier (2006) found a reduction in flux from 20 to 9 Lmh by increasing TSS from 20 to 40 mg/L. Also, flux was negatively correlated with TSS concentration by Dereli et al. (2014) for the SRT of 50 days. However, the aforementioned tendency could not be verified for the SRTs of 20 and 30 days in the same study. This can be related to the lower concentrations of operation on those reactors of 16.5 g/L and 17.2 g/L, respectively, compared to the 28.3 g/L on the 50 days SRT reactor. According to Pollice et al. (2007), there is a threshold value of 20-22 gTSS/L above which the CST of the sludge is significantly influenced by TSS concentration. Therefore, given the MLSS concentration in this reactor, it is not expected to be a limiting factor in terms of sludge filterability.

4.2.6 COD removal efficiency

The COD removal of the system was above 96% for the whole period of study and even higher than 98.5% during the last month of operation. These values are in accordance with the findings of Saddoud et al. (2007) for whey wastewater treated in an AnMBR. However, removal efficiencies for similar wastewater on other high-rate reactors, like UASB or AF are generally around 80 – 95% for stable operation (Karadag et al., 2015a; Karadag et al., 2015b). The main difference is given by the high solids retention efficiency of the ultrafiltration membranes in comparison of no-membrane separation systems.

The high level of COD removal reached for dairy WWs in anaerobic systems compared to other substrates is related to their high biodegradability. Anaerobic biodegradability of dairy WW with similar composition to the one of this study, based more on carbohydrates and proteins than fats, was verified to be above 95% for most concentrations tested (Vidal et al., 2000).

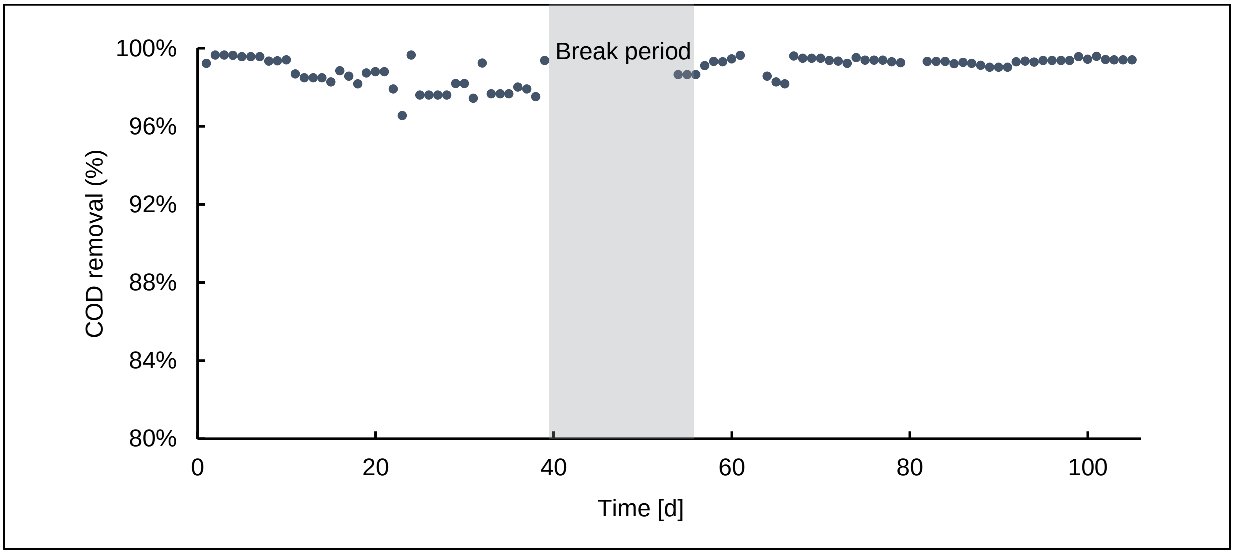


Figure 4-7 COD removal in the AnMBR

4.2.7 COD balance

The results of the COD balance per week are presented in Figure 4-8. It can be noticed from the figure that the cumulative amounts of methane, wasted sludge and COD of the effluent add an average of 86% of the COD fed. The main possible causes for this difference are biogas leakages in the system; biogas lost dissolved in the effluent, higher COD concentration in the effluent during feeding hours, consumption by sulphate reducing bacteria and 0.5L of biogas used 5 times a week for measurement of its composition. In table 4-4 the estimated amounts for the last four factors are presented.

Considering there is still a difference higher than 10%, the rest of the COD is expected to be related to biogas leakages. This is based on the fact that due to the high mixing velocity in the reactor, the mixer axis was identified as the most susceptible point for biogas losses. Even though grease was applied almost daily in that area, leakages were detected throughout all the operation, especially during the feeding period when the biogas production is at its highest rate. Additionally, for days were no leakages were detected, methane represents up to 85% of the total COD fed, and the total COD added is 97-98%.

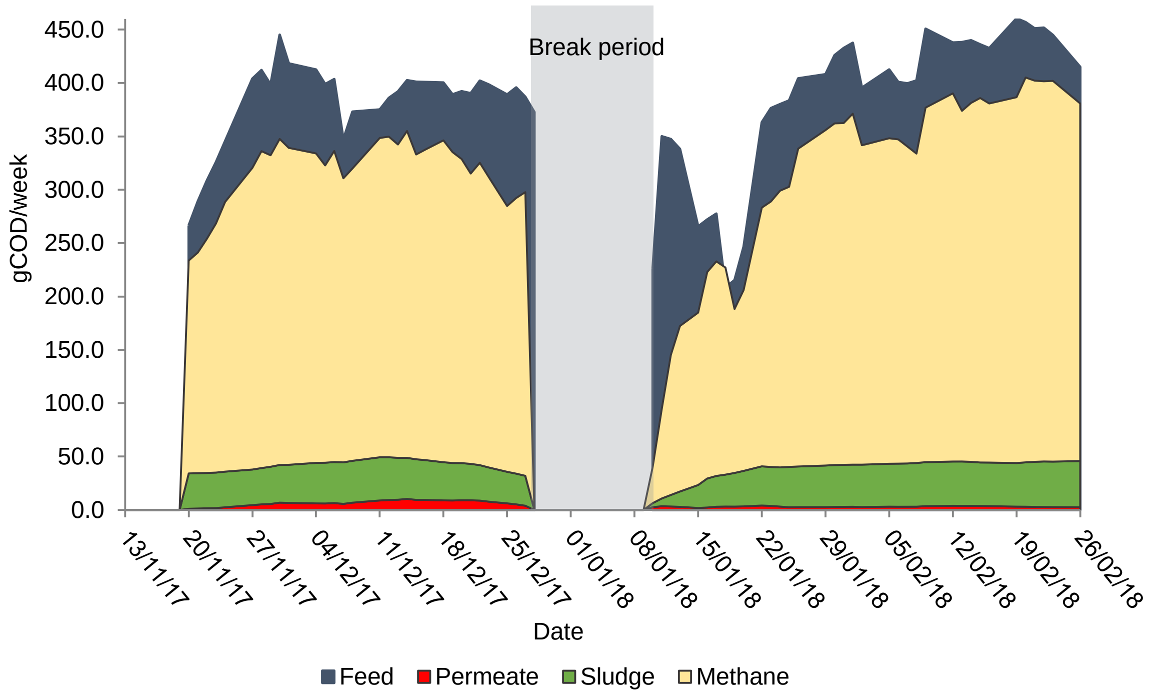


Figure 4-8 Weekly COD balance showing feed and cumulative permeate, sludge and methane

Table 4-4 COD balance summary and possible causes of COD balance differences – last month

Source	**% Feed COD average**	**% Feed COD minimum**	**% Feed COD maximum**
Methane	75	61	93
Sludge	10	8	14
Permeate	0.8	0.6	1.5
Subtotal 1	**86**	-	-
Solubilized CH4 (35^{0}C)	0.4	0.3	0.5
Sulphate reducers	0.6	0.5	0.7
Different permeate COD in cycle	0.4	0	0.7
Biogas for composition measurement	1.5	1.4	1.7
Subtotal 2	**89**	**-**	**-**

Conversion of COD into methane was on average 75 – 80% with the exception of some days were specific biogas leakages were identified in the reactor. These values correspond to an average specific methane production of 0.27 $L_{CH4}/kgCOD_{removed}$. However, if the important biogas losses in the system are considered, for the periods with no leakages, the specific methane production corresponds to 0.30 $L_{CH4}/kgCOD_{removed}$. The obtained results are within the range of dairy wastewater reported values, from 0.28- 0.34 $L_{CH4}/kgCOD_{removed}$. Particularly, for Saddoud et al. (2007), also in an AnMBR the obtained value was 0.30 $L_{CH4}/kgCOD_{removed}$. On the other hand, a higher value of 0.32 $L_{CH4}/kgCOD_{removed}$ was obtained by Gutiérrez et al. (1991) in an UASB reactor treating whey wastewater.

Regarding biomass growth, the observed rate for this experiment of 10% is within the expected values for anaerobic systems that range from 5 up to 20% at high temperatures (Von Sperling and de Lemos Chernicharo, 2005). The relation of COD/VSS was always maintained from 1.5 to 1.7 during the operation. It means some not-degraded COD is being wasted with the sludge considering that in general values for biomass COD content range from 1.42 to 1.48 gCOD/gVSS (Henze et al., 2008).

To determine whether if the accumulated COD was biodegradable or not, batch assays similar to SMA without acetate addition were performed. The total methane production observed during the first 36 hours (no endogenous respiration) corresponded to a concentration of 1.1 gCOD/L (see Appendix C). Subtracting the accumulated COD, the COD/VSS of the sludge results to 1.5 gCOD/gVSS, which is closer to expected values. This shows that higher SRT of the sludge can be used to further stabilize the sludge and increase substrate's methane conversion. However, the already mention drawbacks of operation at higher SRTs for AnMBR should be considered.

4.2.8 Feast-famine cycle study

One complete feast-famine cycle was studied in order to determine variations in permeate COD and VFAs, reactor volume, temperature, pH, biogas production, sludge CST, real flux and TMP of the system (Figure 4-9). The COD and VFA profiles follow the expected pattern, presenting the highest concentrations of 400 mgCOD/L and 3.1 meqVFA/L after the feeding period and being reduced during the fasting. However, even though the highest values are expected to be registered at minute 60 of the cycle, in both cases it was observed at minute 90. This could be due to the volume of permeate accumulated in the membrane tube which acts as a "buffer" volume and therefore the permeate effluent at a specific moment is not exactly the one filtered at that instant (concentration in the reactor).

In the 4 hours following the feeding period, both COD and VFA concentrations reached similar levels to the ones at the start, 113-130 mgCOD/L and 0.3 – 0 meqVFA/L, respectively. Also, the biogas production registered during the last 2 hours of the cycle was very low meaning that most of the COD degradation was completed in 2-3 hours from feeding. More studies should be done in order to optimize the fast-feeding durations as decreasing fasting time for full-scale might mean savings on buffer tank volumes.

The pH, temperature and volume profiles also follow the predictable tendency. One factor important to observe is that the minimum pH which was reached at the end of the feeding period was 6.7. In this case, the pH of the feed was 4 and taking into account the amount fed (0.82 L), the pH could have dropped up to a value of 5.2 if not for the alkalinity present in the system. This is also in accordance with minimum pH values of 6.5 registered when pH of the feed was at a minimum of 3.6.

Regarding sludge filterability properties, the measured CST during the cycle could not be directly related to the TSS concentration. This can be due to the already mentioned fact that only above a limit concentration, the effect of MLSS on CST is made clear. However, in general during the last 30 minutes of feeding and up to 1 hour after feeding the TMP registered was lower while the real permeate flux measured slightly higher. There might be some relation between these results and the “dilution” of the sludge, but results obtained are not conclusive.

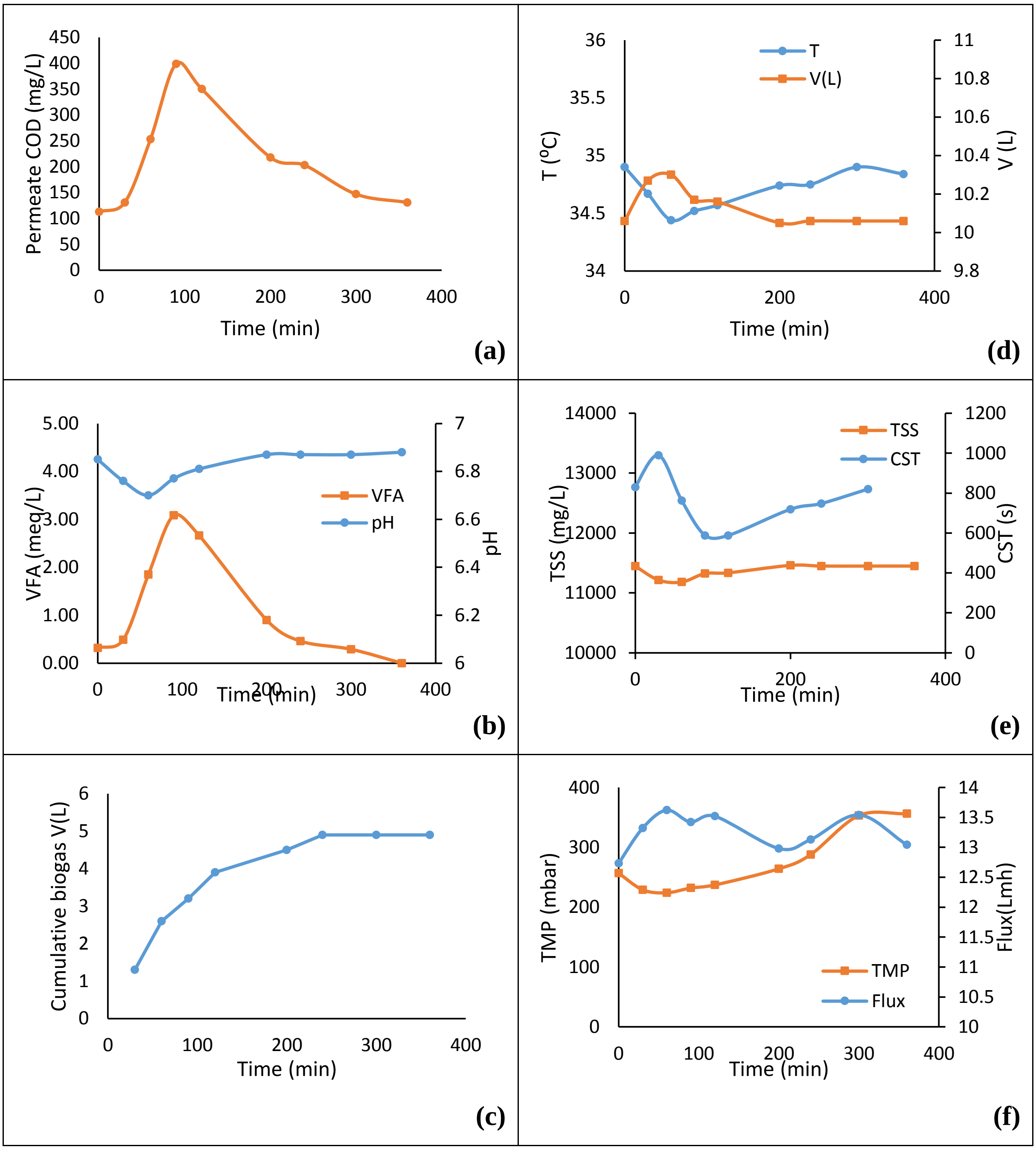


Figure 4-9 Fast-feeding cycle profiles

(a)Permeate COD (b)VFAs concentration and reactor pH (c)Biogas production (d)Reactor volume and temperature (e)CST and TSS of sludge (f) TMP and real flux measured

4.3 Sludge filterability characteristics

Several researchers tried to relate properties of the sludge suspension with the maximum obtainable flux in a system. Suspended solids concentration, already discussed in section 4.2.5, has been directly related to membrane fouling propensity in several studies (Jeison and van Lier, 2006; Pollice et al., 2007). However, some contradictory results showed that in order to assess real fouling capacity of the sludge, other parameters should also be considered for a more accurate indication (Le-Clech et al., 2006). Many different suspension characteristics have been used in the abovementioned studies, from which the ones chosen for this study were: capillary suction time (CST), viscosity, particle size distribution, morphology and relative hydrophobicity. SEM/EDS analyses were additionally used in order to evaluate the presence of inorganic precipitants in the sludge.

4.3.1 CST and Viscosity

For this MSc research, a reactor that had been running for more than 400 days was taken over, and the influent was changed from diluted milk to real dairy WW. During the whole period of operation of this reactor, CST and viscosity were measured and were highly correlated (Pearson correlation coefficient: 0.85 and P value: 0.0001), therefore they are going to be discussed concomitantly.

It was based on results of these parameters that the reactor feeding was changed from continuous to intermittent during the phase of synthethic feeding. After the 3 SRTs of operation, a high increase in CST and viscosity values from 20-100 s and 5 – 7 mPas, to 1500 s and 15 mPas, respectively, were observed (Figure 4-10). Additionally, the operational TMP of the membrane increased from 100-200 mBar to 600 mBar. Therefore, these were the main drivers for changing the feeding regime in an attempt to improve the sludge filterability quality. This was actually efficient as during the operation with synthetic WW in intermittent feeding values of CST and viscosity ranged between 400 – 800 s and 5 – 9 mPas, respectively (Figure 4-10). However, the effectiveness on the filtration performance could not be proven as the TMP was highly variable ranging from 300 to 750 mbar while the flux was generally constant around 11-12 Lmh (see Appendix D for previous operation flux and TMP).

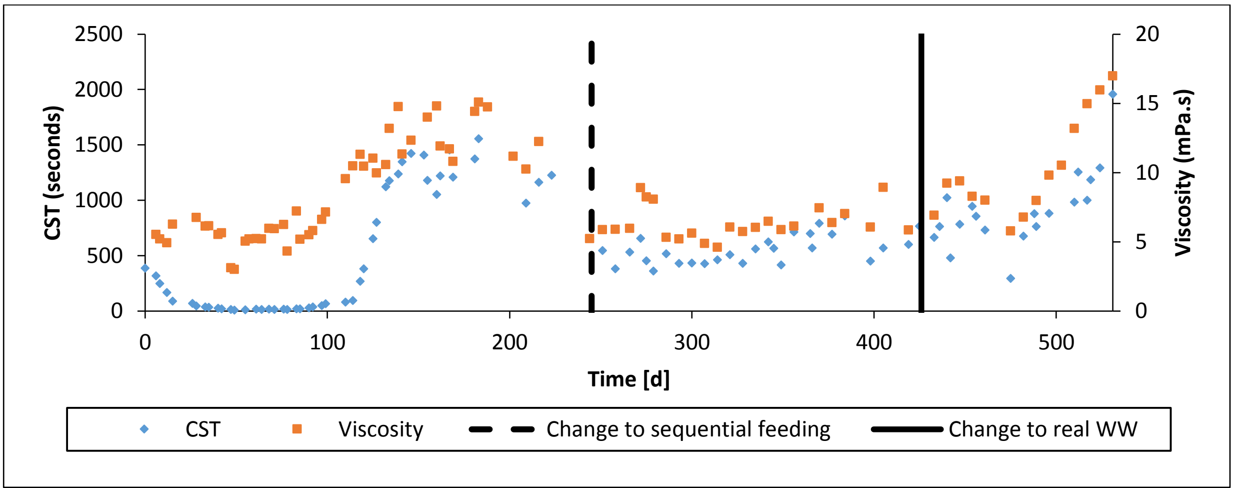


Figure 4-10 CST and Viscosity of sludge during the complete period of reactor operation

On the other hand, both CST and viscosity started to increase after changing the substrate from synthetic to real dairy WW, especially after 2 SRTs completed. At the end of this study, as can be seen in Figure 4-10, the correspondent values for CST and viscosity reached 1960 s and 17 mPas, respectively, being even higher than under continuous feeding with synthetic WW (Figure 4-10). However, it should be noticed that, despite the poor sludge quality, after chemical cleaning of the membrane, the obtained flux and TMP at the end of the period resulted of around 70 mbar for a flux of 13 – 14 Lmh, respectively, representing the highest sludge permeability after 1 year of operation (see section 4.4.1).

The obtained CST values are in the same range as the ones reported by Dereli et al. (2014) for the treatment of stillage from corn-based ethanol production. Different SRTs of 20, 30 and 50 days were applied in that study, reporting average CSTs of 951 s, 1743 s and 2414 s respectively. However, CST was proven to be correlated with TSS concentration (Dereli et al., 2014; Pollice et al., 2008); therefore, normalization of the values should be used for comparison. In this case, the average normalized values were 61, 90 and 86 sL/gTSS for the SRTs of 20, 30 and 50 days respectively. The average normalized CST for this study was 87 sL/gTSS, which is similar to the aforementioned research but at the end of the operation, a value up to 150 sL/gTSS was measured. Sludge conditions should be further followed-up as probably, the steady-state value for the system has not been reached yet.

An important observation that was made from the last period of operation with real wastewater was that the increase in CST and viscosity started particularly after stopping the control of reactor pH. Both parameters were plotted against pH (Figure 4-11) to identify the existence of any particular effect. Pearson correlation coefficients were -0.47 and -0.64 for CST and viscosity respectively indicating a significant negative correlation with 95% confidence level (p-values: 0.04 and 0.005).

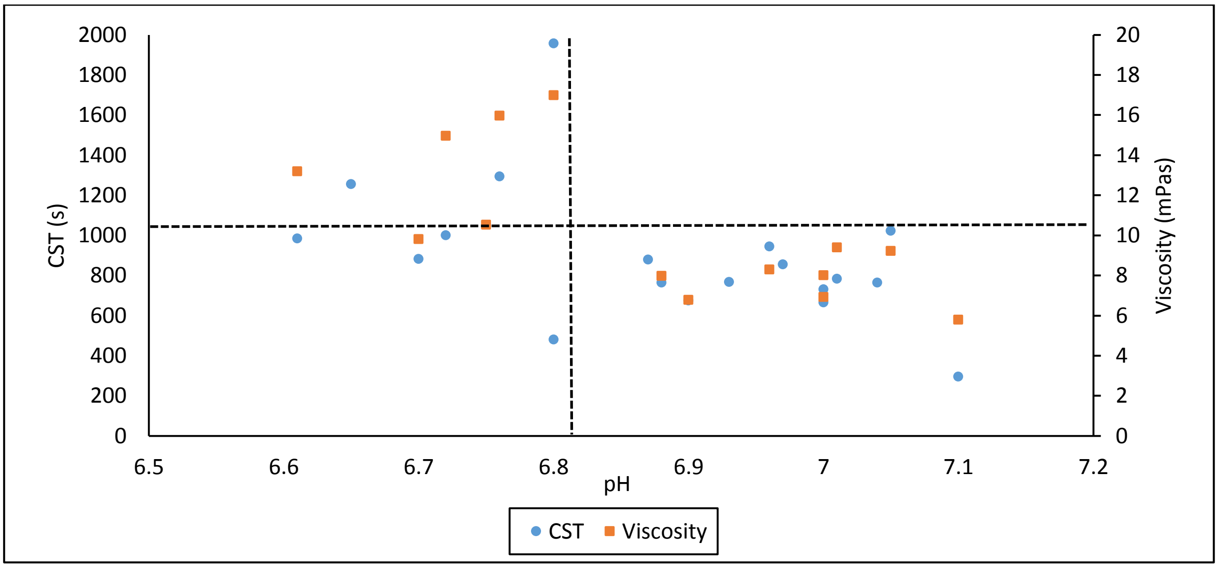


Figure 4-11 Correlation of CST and viscosity with reactor pH

Different factors can explain the effect of the operational pH on these filterability properties. Aquino and Stuckey (2004) reported that a pH reduction in a system from normal operation at 7 to values up to 6.2 increased SMP concentrations presumably in relation to fermentative and acidogenic bacteria growth. A similar situation occurred in the reactor of this study but in a narrower range. Given the low alkalinity of the feed, while operating without base addition, throughout feeding hours the pH dropped up to 6.5 from the average level of 6.8. Additionally, Ghaly (1996) reported that lower operational pH promotes filamentous bacterial growth. Long filaments were observed in the morphology study performed by KU Leuven and also in SEM images of the sludge (see sections 4.33 and 4.3.4). The presence of filamentous bacteria was related to severe fouling in aerobic MBR due to the formation of a non-porous gel layer (Meng et al., 2006). Moreover an increase in viscosity of the sludge was observed with the increase of filamentous bacteria in the study of Meng et al. (2007) presumably due to EPS accumulation.

4.3.2 Particle size distribution

The particle size of the sludge was measured 6 times during the total operation of the reactor. Only one measurement is available for the first 2 months of operation while the other 5 were done weekly in the last month of operation to assess the sludge condition during membrane evaluation. Results of the particle size distribution for the sludge are shown in Figure 4-12.

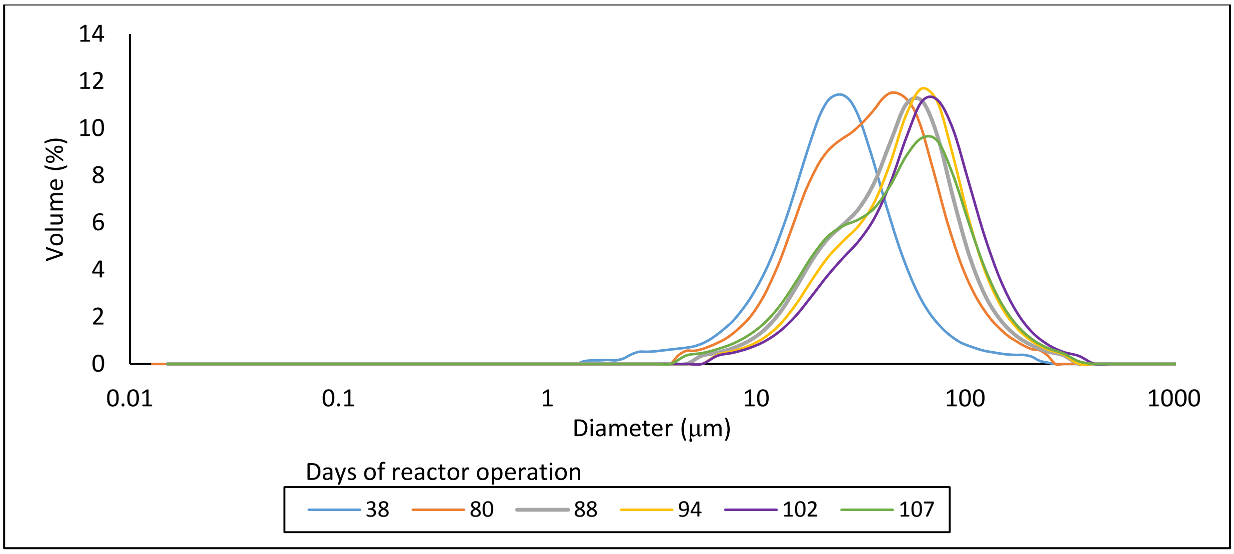


Figure 4-12 Particle size distribution during different days of reactor operation

A general increasing tendency on the mean size was observed from day 38 of operation to day 102 as can be also be noticed from mean size values and percentiles 10, 50 and 90 shown in table 4-5. This tendency is opposite to the generally reported decreased in particle size in cross-flow configurations due to the high shear effect of pumping (Kim et al., 2001; Le-Clech et al., 2006). It is important to note that in those studies mainly centrifuge pumps were used. In this study, the recirculation pump of the reactor was an eccentric screw pump which operates with lower shear stress. However, the last measurement on day 107 presented lower mean values than day 94 despite no changes were done in the reactor operation.

Table 4-5 Summary of particle size distribution measurements

Day	38	80	88	94	102	107
d10 (μm)	9.75	13.18	16.29	17.91	19.75	14.78
d50 (μm)	23.07	35.03	45.23	50.46	56.15	46.63
d90 (μm)	50.57	83.38	95.27	104.70	119.17	107.77
MV (μm)	28.50	43.89	52.74	57.96	65.31	56.11

From the results of this study a positive correlation between median particle size and CST of the sludge is suggested. However, from the first operational period of this AnMBR with synthetic wastewater, an opposite tendency was observed (Figure 4-13). Sethi and Wiesner (1997) modelled cross-flow ultrafiltration determining that small particles in the range of 0.4 µm induced a decrease in permeate flux. Likewise, Kim et al. (2002) verified that small particles control the cake formation due to their higher tendency for deposition thus affecting the filterability. Although the increase measured in the mean particle size the actual number of small particles could remain the same, and therefore the filterability of the sludge is not correlated with this parameter. Additionally, as discussed in the previous section, filaments presence might increase particle size and fouling at the same time.

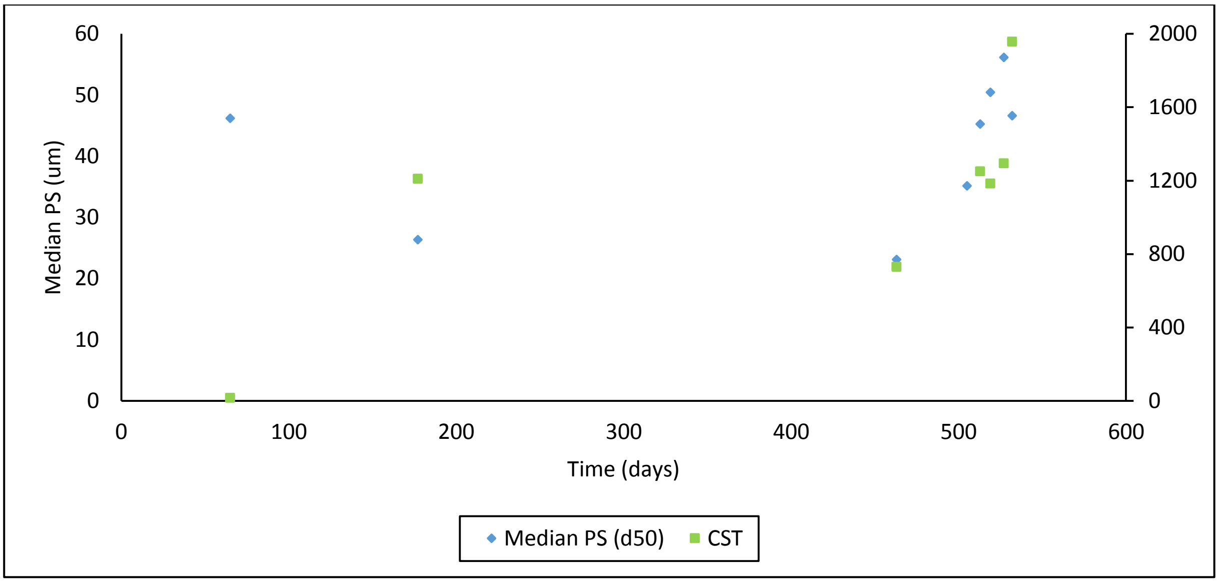
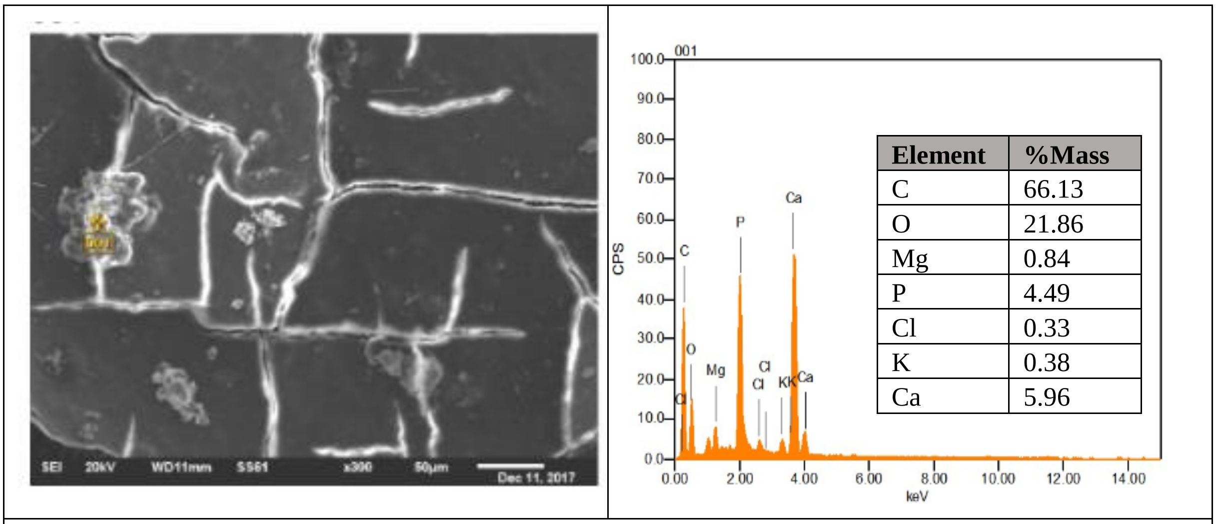


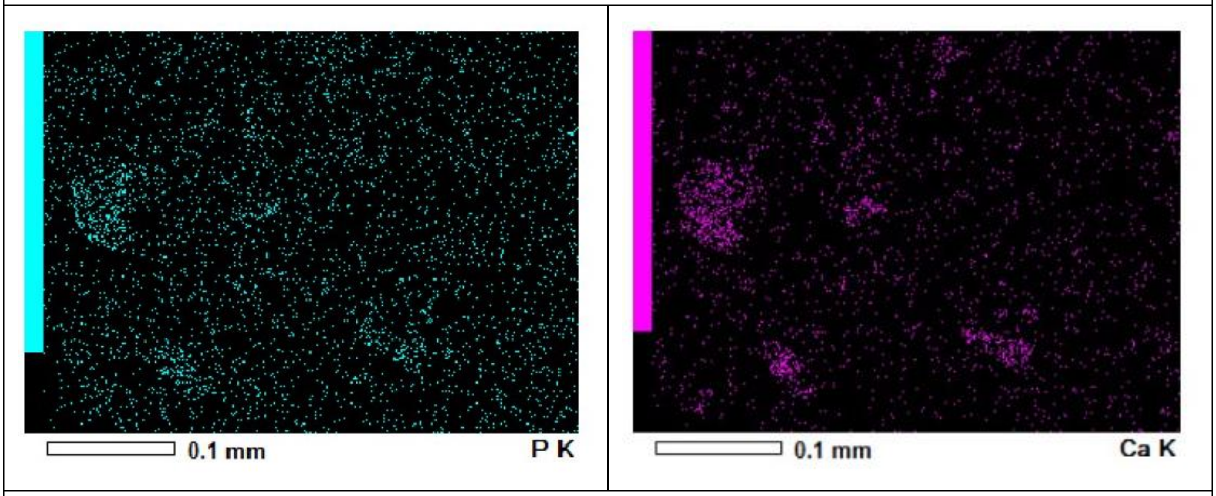
Figure 4-13 Median particle size and CST overall reactor operation

4.3.3 SEM images and EDS for the study of the elemental composition of precipitates

During the first month of operation using real dairy, WW precipitates accumulation on the sludge was observed, resulting in a big failure in the system due to clogging of the pump's suction site. The composition of samples of precipitates and dried sludge collected from the reactor in December and January were analysed using SEM/EDS. Calcium and phosphorous were identified as the main components of amorphous inorganic structures observed in the samples as it is shown in Figures 4-14 and 4-15. Also magnesium but in lower proportion was found. This is in accordance with the higher calcium concentrations compared to magnesium measured in this particular substrate and expected from dairy wastewater.



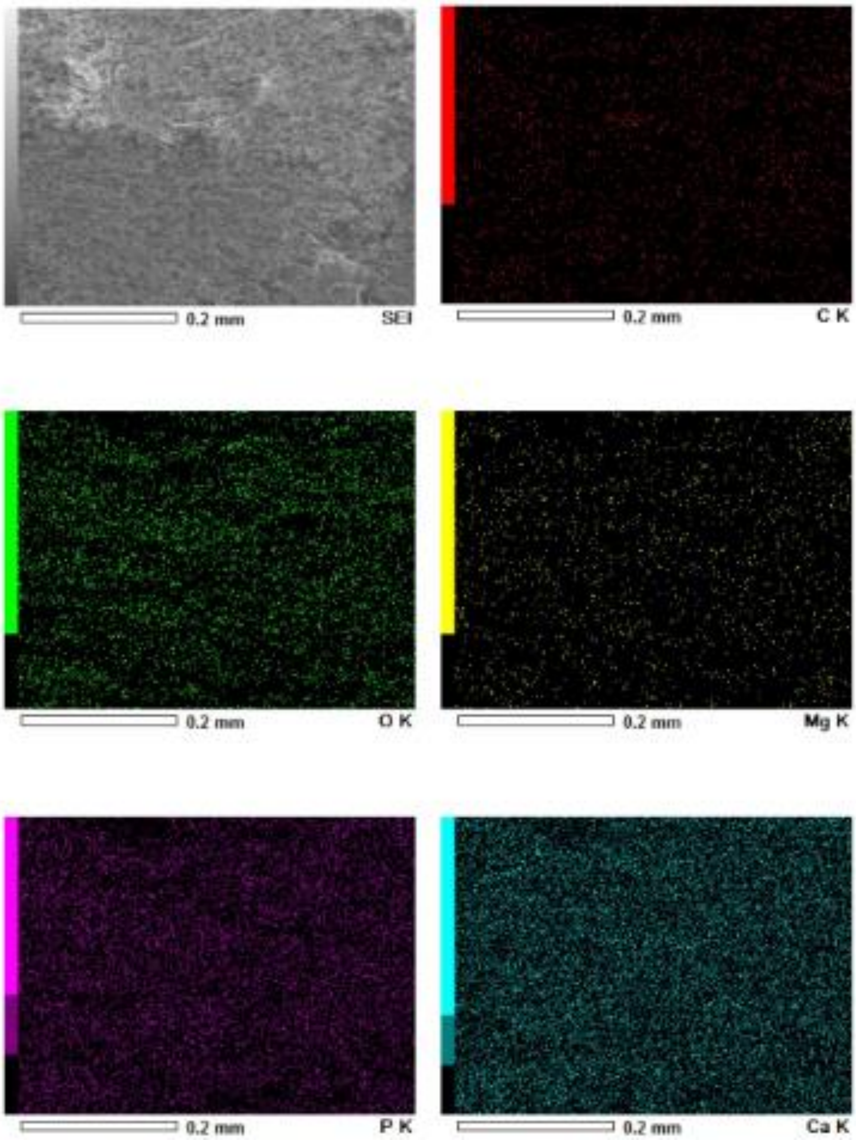
Elemental analysis of selected point in an amorphous inorganic structure

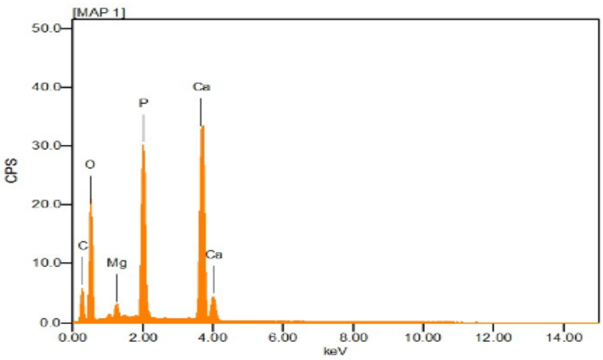


Mapping results of the same image showing Ca and P concentration areas

Figure 4-14 Results of first EDS analysis of precipitates on 11/12/17 (day 28)

Precipitants like struvite, calcium phosphates or calcium carbonate are more prone to cause inorganic fouling in anaerobic reactors than in aerobic systems due to the higher concentrations of ammonium and phosphate and the pH variability given by CO_2 presence (Liao et al., 2006). All those compounds have been found in the membrane fouling layers of different AnMBRs (Jun et al., 2017; Zhang et al., 2007). After cleaning of the reactor for removal of precipiatants, elemental analysis of sample showed mainly calcium phosphate (Figure 4-15). Given that solubility of all calcium phosphates increases by decreasing pH (Chow, 2001), as it was already explained, the addition of base was stopped aiming at reducing the number of precipitants. Moreover, as will be further explained in section 4.4.2, membrane autopsy performed by the supplier after it was changed on day 22 revealed a high level of damage on the pores probably due to erosion caused by the inorganics precipitants.





Element	%Mass
C	30.14
O	52.22
Mg	0.74
P	7.72
Ca	9.18

Figure 4-15 Mapping elemental analysis of a sample collected from reactor interior cleaning showing high presence of Ca and P – 03/01/18

Concentrations of anions and cations in the feed and permeate of the reactor were evaluated and they are shown in Figure 4-16 for the operation at average pH of 7.0 and 6.8. A similar reduction in the concentration of both Ca and phosphate was observed for both conditions. It should be noted that higher concentration of phosphorus in the wastewater was measured during the operational period at pH 6.8 and it was related to a different batch of wastewater. That resulted in a higher P concentration in the permeate, but the amount of phosphorus removed by precipitation and growth remain the same.

However, from the direct observation of the sludge and SEM/EDS analysis on the operation at a pH of 6.8, less presence of big size inorganics precipitates was noticed compared to the sludge at pH of 7. The results of mapping on the sample of sludge collected at pH of 6.8, presented in Figure 4-17, showed that the elements were still present but mainly within biological formations. This can also be in relation with the observed increase in particle size of the reactor as calcium is well-known as a promoter of bio-flocculation for anaerobic reactors (Pavoni et al., 1972). In fact, You et al. (2005) proved that entrapment of inorganics in biological formations is efficient for the reduction of membrane scaling.

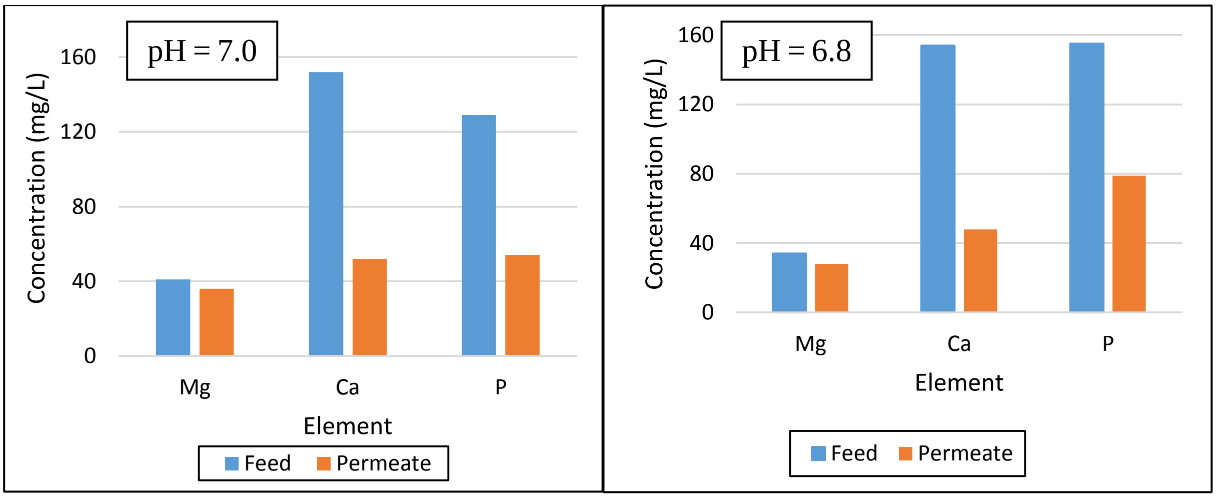
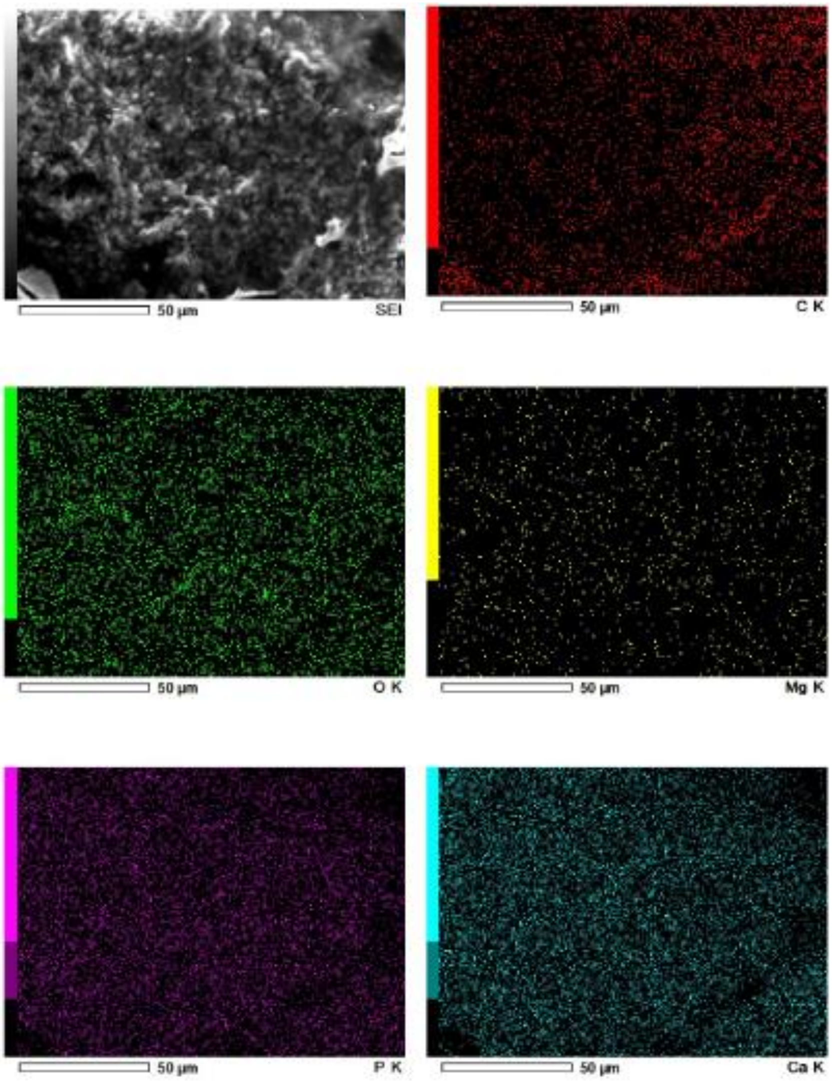
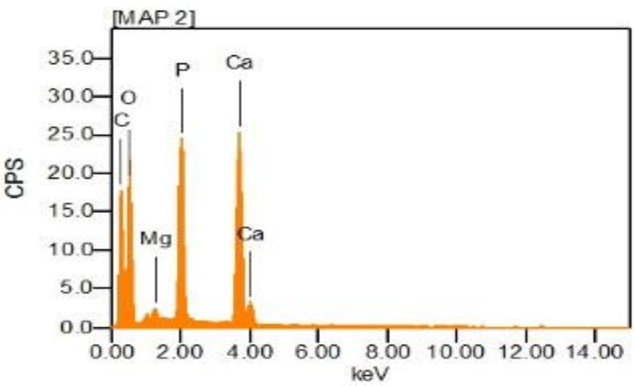


Figure 4-16 Concentrations of Mg, Ca and P in feed and effluent at pH of 7 and 6.8





Element	%Mass
C	51.75
O	38.76
Mg	0.35
P	4.38
Ca	4.76

Figure 4-17 Mapping elemental analysis of a precipitate sample collected from the reactor – 21/02/18

4.3.4 Morphology of sludge

As part of the PhD research of Pieter Van Gaelen at KU Leuven University, different analyses were done on sludge samples were taken every 2 weeks. Morphology results indicate average floc size started at about 30 µm (Figure 4-18) and reached up to 50 µm (Figure 4-19). These results are in accordance with observations of particle size distribution presented in section 4.3.2. Additionally, a large number of small fragments was identified in the images which can be the cause of the high CST measured as was explained in section 4.3.1.

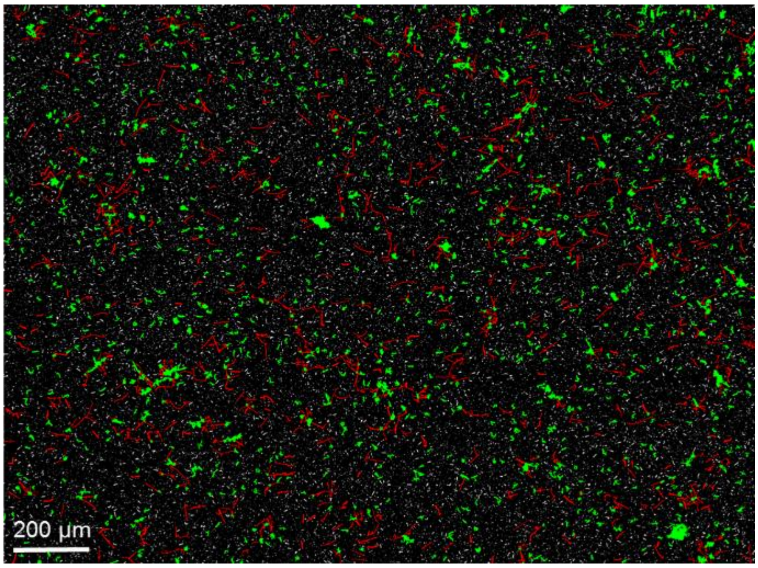


Figure 4-18 Sludge dilution to 1 mg/L image after software contrast – Synthetic feeding. Source: Van Gaelen (2018) Code: Green – Flocs (>5um), Red-filaments, White-Fragments (<5um)

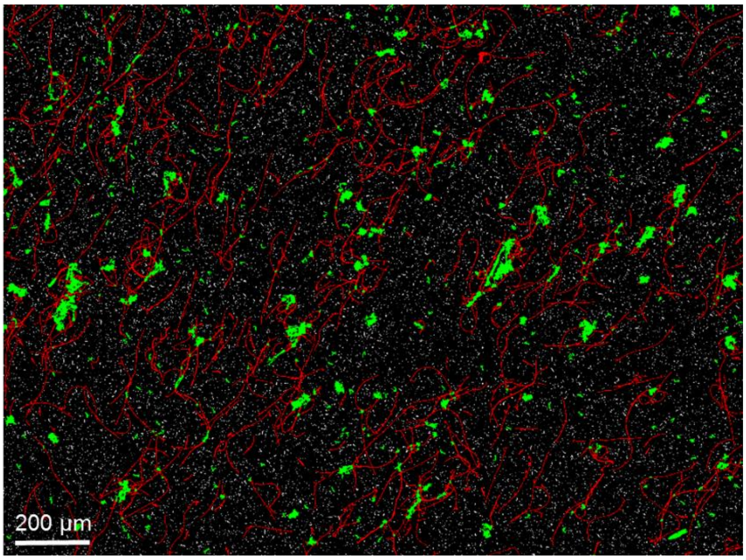


Figure 4-19 Sludge dilution to 1 mg/L image after software contrast – Dairy WW. Source: Van Gaelen (2018) Code: Green – Flocs (>5um), Red-filaments, White-Fragments (<5um)

It can also be noticed when comparing figures 4-18 and 4-19 that the change of feeding from diluted milk to real dairy WW correlated with an increase in the number of filaments. This is in line, as mentioned in the previous section with the poorer dewaterability characteristics of sludge. Additionally, in the last analysis of sludge with SEM, even though gold coating of the sludge was not possible due to its high humidity content, presence of filaments was observed as it is shown in Figure 4-20.

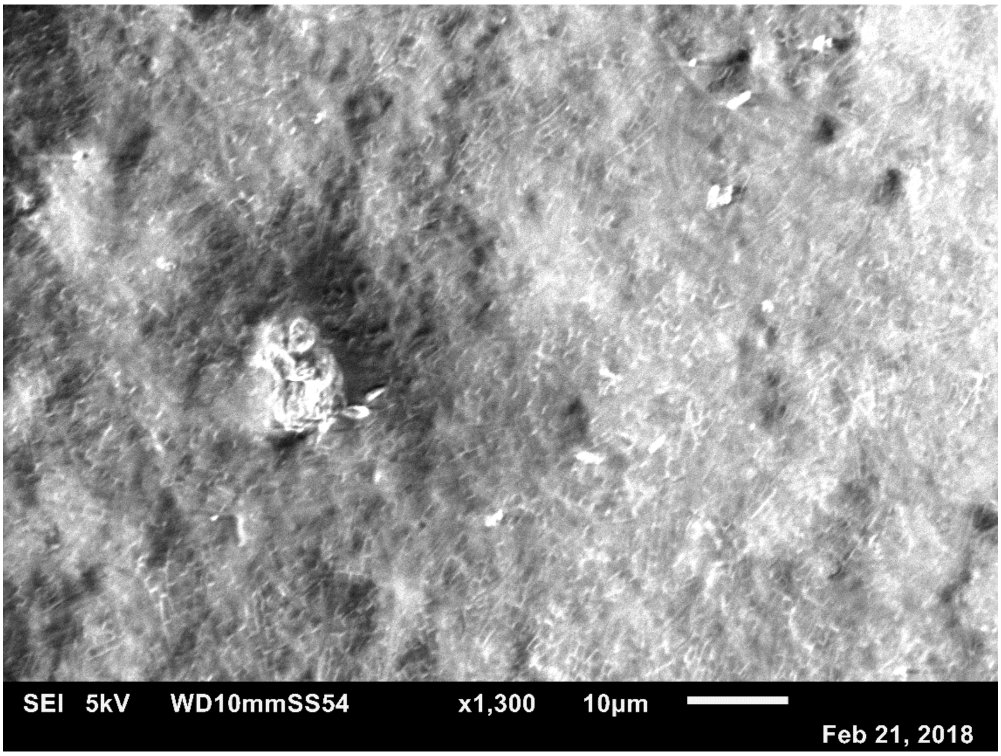


Figure 4-20 SEM image of sludge on day 100 of operation

4.3.5 Relative Hydrophobicity

Relative hydrophobicity (RH) is related to different characteristics of the sludge as EPS, microbial community or substrate accumulation (Dereli et al., 2015).This parameter was also measured in the context of the PhD of Pieter Van Gaelen. Results of RH are presented in Figure 4-20 for the period of operation of the reactor in this study. As it can be seen no significant changes on RH were noticed with time. On day 56 the sample was taken after opening and cleaning the reactor so it might not be completely representative.

The influence of relative hydrophobicity on membrane fouling is still not clear as contradictory results have been found. In the study of Van den Broeck et al. (2011) high RH values from 40 up to 90% were measured for activated sludge, but the higher values did not correspond with the lower sludge filterability suggesting that this factor alone is not enough to predict filtration characteristics. On the other hand, in the study by Meng et al. (2006) the higher the values of RH, the higher the EPS measured and consequently the higher the membrane fouling. In that study it was related to a higher adherence of the flocs to themselves and also to the hydrophobic membrane, which can be similar in this research.

In the treatment of high-lipid-content wastewater, values from 34 to 58% were measured by Dereli et al. (2015) relating the highest value with high LCFA accumulation. In that study, the high sludge RH was found a relevant factor to hinder membrane fouling. However, the FOG concentration in that substrate was more than 10 times higher than the one measured for the dairy WW used in the present study, so this factor is not expected to be dominant in the present study. On the other hand, also EPS concentrations can lead to high RH, and the presence of filaments has been related to high EPS levels (Meng et al., 2006). This factor is more prone to be the controlling condition in this reactor.

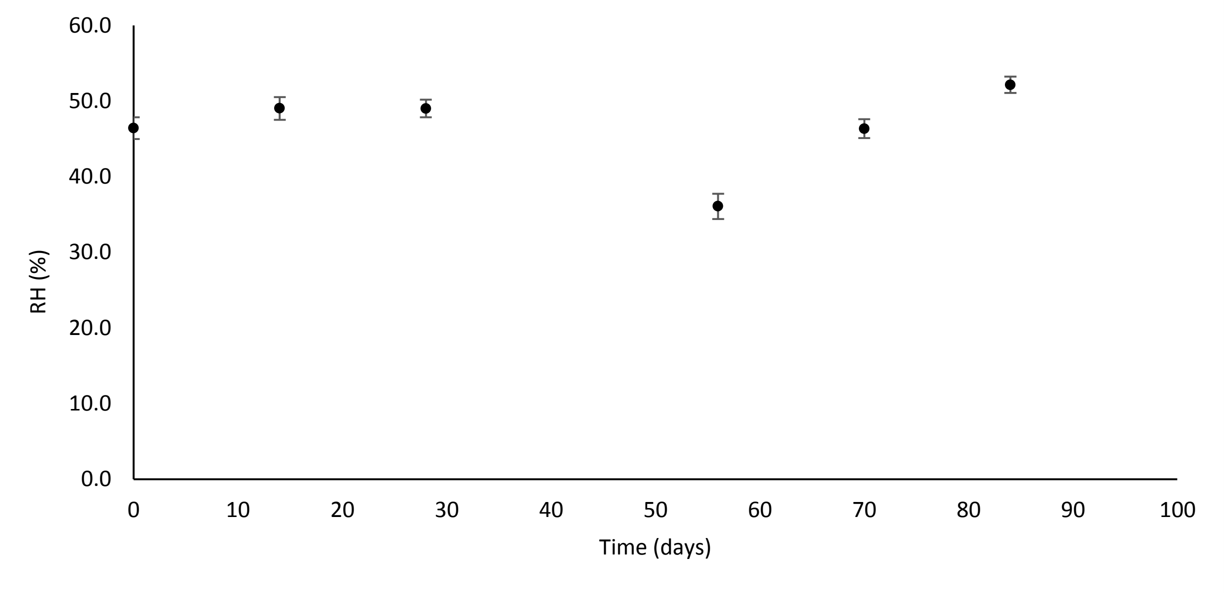


Figure 4-21 Relative hydrophobicity of the sludge. Source: Van Gaelen (2018)

4.4 Membrane performance

4.4.1 Flux and transmembrane pressure

Real flux in the system was measured daily to verify behaviour with respect to setpoint. Despite efforts on the calibration of the permeate pump to adjust real permeate flux to the value set on the PLC, it was not possible to reach the desire flux values controlling the system with the permeate pump. This can be noticed in the analysis of Figure 4-22 were setpoint and real flux are plotted, and no correlation between them is observed. Additionally, from Figure 4-23 where the average TMP registered during each real flux measurement is shown, the absence of correspondence between TMP and flux in this system is also clear. Other fact that can be derived from the comparison of these two figures is that no significant changes were obtained after replacing the membrane for a new one, while the CIP done on day 100 had a positive effect on membrane performance (see section 4.4.2).

The average values of flux and TMP throughout reactor operation were 12 Lmh and 400 mbar respectively, corresponding to an average permeability of 30 Lmh/bar. A similar permeability but a lower flux of 9 Lmh was reported for treatment of high lipid-containing WW (11.3 gFOG/L) also at an SRT of 30 days and similar sludge characteristics (MLSS, CST and mean particle size) but at different filtration parameters, cross-flow velocity of 0.5 m/s and cycles of 30 s backwash every 300 s of filtration (Dereli et al., 2014; Dereli et al., 2015).

However, similar full-scale installations from Biothane® treating dairy wastewater operate at higher fluxes and TMP. The correspondent ranges observed are 20 – 25 Lmh and 100-200 mbar, resulting in up to 4 times higher permeability (Bouman and Heffernan, 2010). As a consequence, membrane performance observed in laboratory reactors is still not considered trustworthy for industrial designs. Scaling down the treatment might affect sludge characteristics due to a higher shear rate as recirculation is 180 and 24 times a day in lab-scale and full-scale, respectively. Nonetheless, for instance, CSTs up to 2000s are also observed in industrial reactors with higher fluxes than the obtained in this study. Consequently, also the differences in the set-up might be causing the unreliable membrane performance.

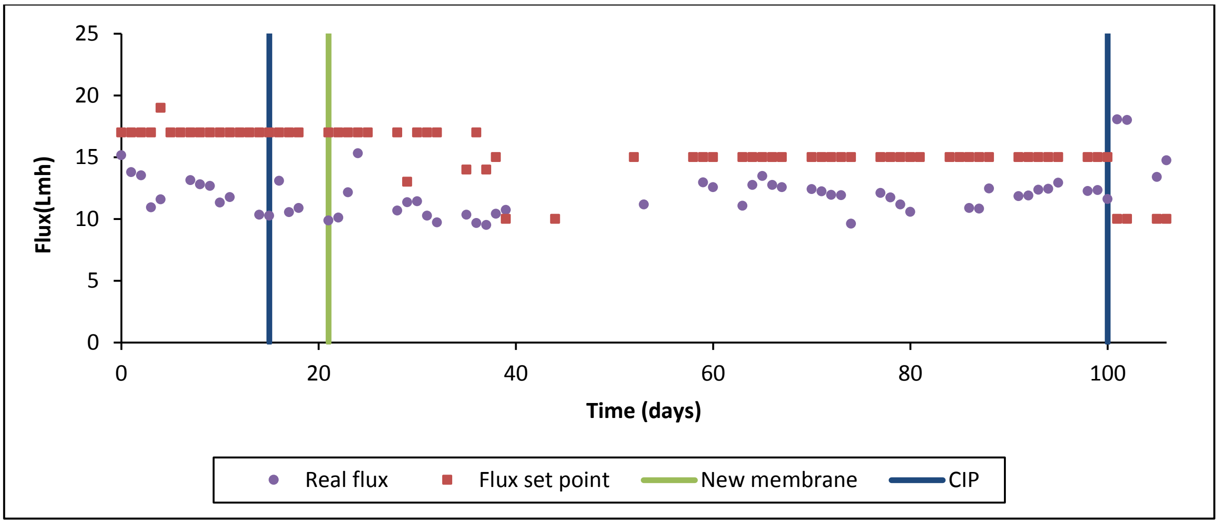


Figure 4-22 Flux set point and real flux over operation

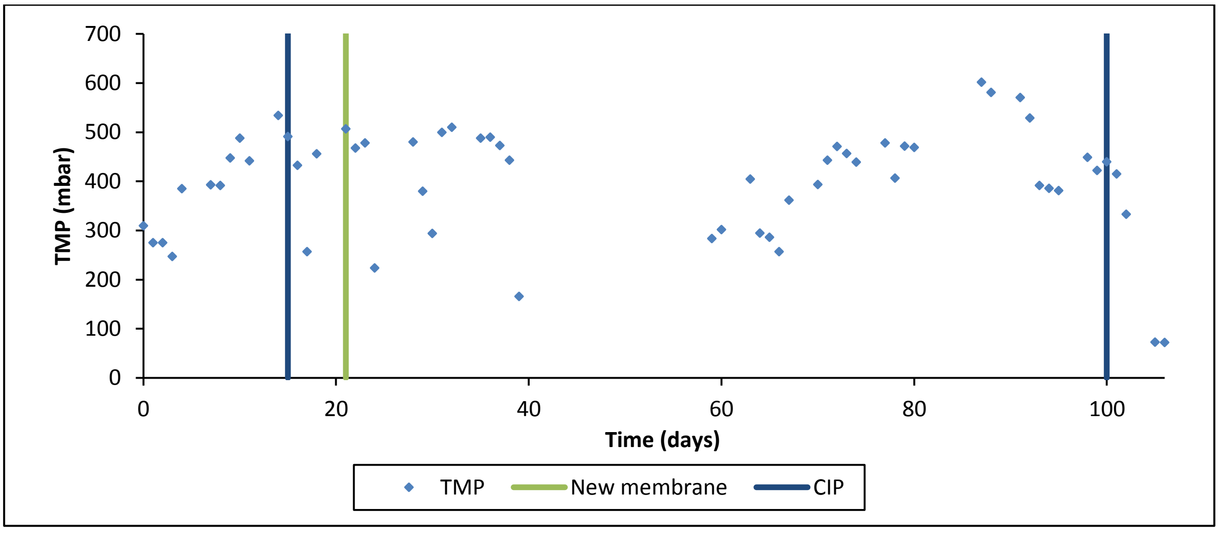


Figure 4-23 Average TMP values during real flux measurement

Particularly in this study values of flux above the median (up to 18 Lmh) were unexpectedly observed when the system was operating at lower TMP. That decrease in TMP was caused by an increase in permeate line pressure due to the displacement of the tubing in the pump. In fact, as it is made clear in Figure 4-24, it is the permeate pressure that controlled the TMP of the system considering that feed and retentate pressures were generally stable.

After analysing the cause of obtaining higher fluxes at higher pressures in the permeate line, while it was expected to be the opposite, it was noticed that bubbles were filling the line instead of liquid when the suction made by the pump was higher (lower pressures). This fact can be linked to the faster formation of the cake layer due to the suction and therefore, the higher the setpoint of flux in the system, the higher the rpm of pump operation, the lower the permeability observed and eventually also the lower the flux. This deficiency noticed in the existent set-up lead to the analysis that will be presented in section 4.5.4.

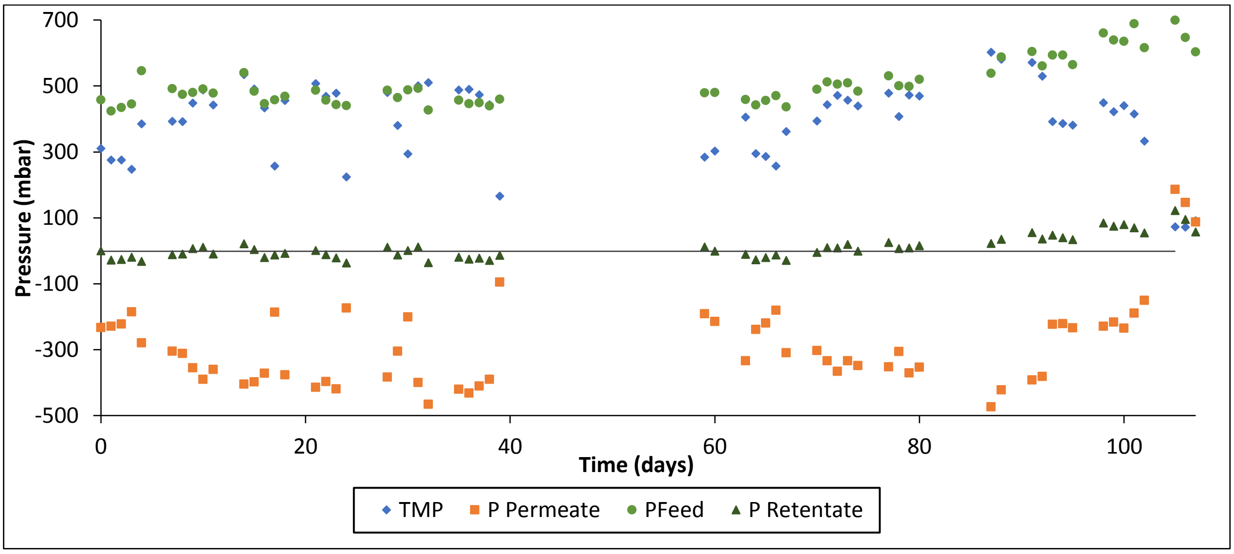


Figure 4-24 Membrane pressures evolution during the operation

4.4.2 Membrane cleaning and CWP

During the 107 days of operation, a total of 2 chemical cleaning cycles were done on the membrane. Given that no improvement on CWP of the membrane was observed after the CIP of day 15, it was decided to change the membrane for a new one in order to avoid the bad membrane condition to affect the performance evaluation. It can be noticed, that even after replacing the membrane, no significant changes on permeate flux were observed. This showed that membrane condition was not the limiting factor in filtration performance at that point, but the set-up itself and the sludge conditions as mentioned in the previous sections were controlling.

The cycles of membrane cleaning included: rinsing with clean water, organic cleaning with HClO and inorganics removal with critic acid in that particular order. The concentration used for both chemicals was 1% as recommended by the membrane supplier (Niejman, 2014). Duration was based mainly on chemical consumption determined by HClO test paper and pHmeter for the different chemicals used. The soaking time resulted in 1.5 – 2 hours for HClO and 3 hours for the citric acid. Results of CWP of the membrane measured at 30°C and a set point of 50 Lmh on permeate pump, before and after every chemical cycle are summarized in Table 4-6 as well as CWP of virgin membranes. Additionally, the correspondent resistances were calculated following Equation 2-1 and are displayed in the same table.

Table 4-6 Clean water permeability during CIP

CIP date	Virgin membrane		Before CIP		After chlorine		After acid	
	CWP	R	CWP	R	CWP	R	CWP	R
Day 15	1000*	0.45	23	198	188	24	203	22
Day 100	4500	0.1	53	85.6	1054	4.32	1726	2.62

() CWP in Lmh/Bar & R in $10^{14} m^{-1}$

*Value suggested by the supplier, not possible to test on the particular membrane as it was already in use

The fractions of each individual fouling source on the total resistance of the membrane are presented in Figure 4-25 as calculated following Equation 2.1 presented in Meng et al. (2009). It should be mentioned that membrane "rinsing" circulating clean water for 15 minutes before starting chemical cleaning can be considered already a physical cleaning step of the membrane and therefore the membrane resistance before CIP could be even higher than the measured values. However, given the short duration of the physical cleaning, it is probable that part of the cake layer resistance is removed together with the organic fouling and therefore included in that resistance.

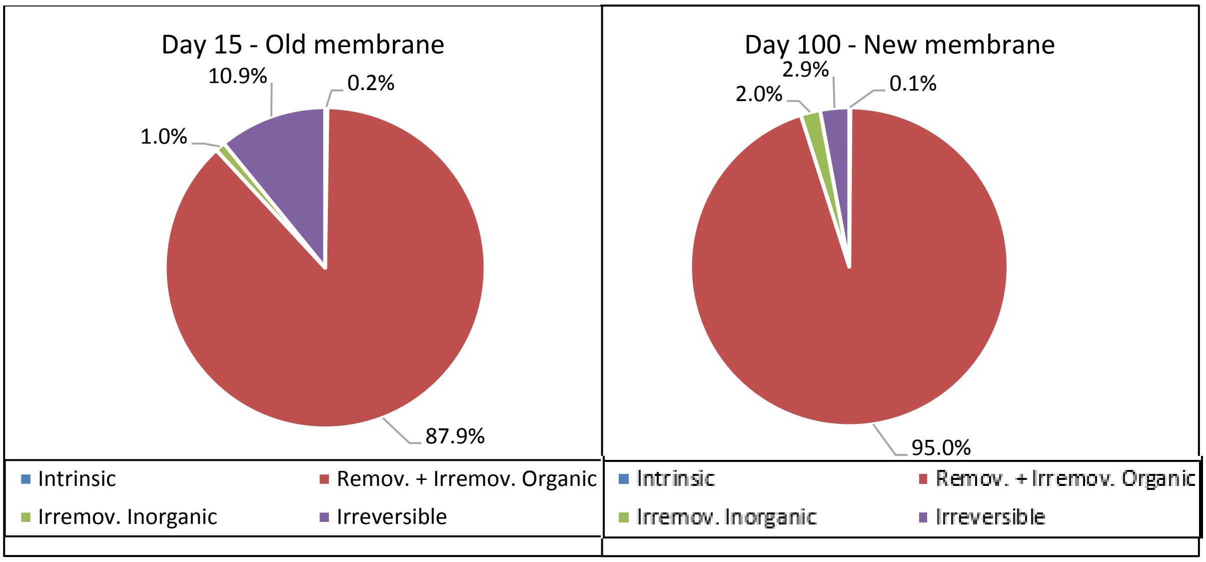


Figure 4-25 Fouling resistances measured during CIP of days 15 and 100

From the analysis of resistances, it follows that in both cases the cake formation and organic irremovable fouling add most of the total membrane resistance. The higher influence of the cake layer is in accordance with reported results of Jeison and van Lier (2006) where it was responsible for 90% of the total resistance. It should be noticed that clear distinction between removable (physical) fouling and irremovable fouling was possible in that study as physical cleaning procedure included the use of jet pressurized water and membrane surface scrubbing.

Despite the aforementioned high presence of crystals in the reactor, the contribution of inorganic fouling for both cases represented a very small fraction of the total membrane resistance. Jun et al. (2017) studied bulk and inside-pore crystallization defining that hydroxyapatite (Ca and P) precipitates almost completely in the liquid while dolomite and struvite (both containing Mg) are more prone to crystallize in membrane pores. Therefore, the low levels of inorganic fouling found in this study can be also related to the low Mg concentration of the substrate avoiding the formation of those minerals. Additionally, inorganics can precipitate within the biofilm, favouring its attachment (He et al., 2005). Therefore, part of the inorganics that were part of the fouling layer might have been attached to organics and removed in the cleaning step with NaClO adding to the irremovable organic resistance.

Regarding the irremovable fouling, the resistance fraction in the old membrane is higher than in the new one, as it was expected considering the operation for more than 1 year of the former one. This was also observed in the autopsy of the membrane executed by the supplier after cleaning by overnight soaking in NaClO. A discoloured look was observed, and SEM images of the surface showed a high level of erosion (Figure 4-26) and some damaged spots (Figure 4-27). Additionally, the fouling layer elemental composition was determined by EDX finding N and O as the main constituents (Table 4-7) suggesting the presence of proteins. These compounds are generally the prevailing part of EPS and have a higher tendency to accumulate in the membrane pores than carbohydrates (Malamis and Andreadakis, 2009; Wu et al., 2008).

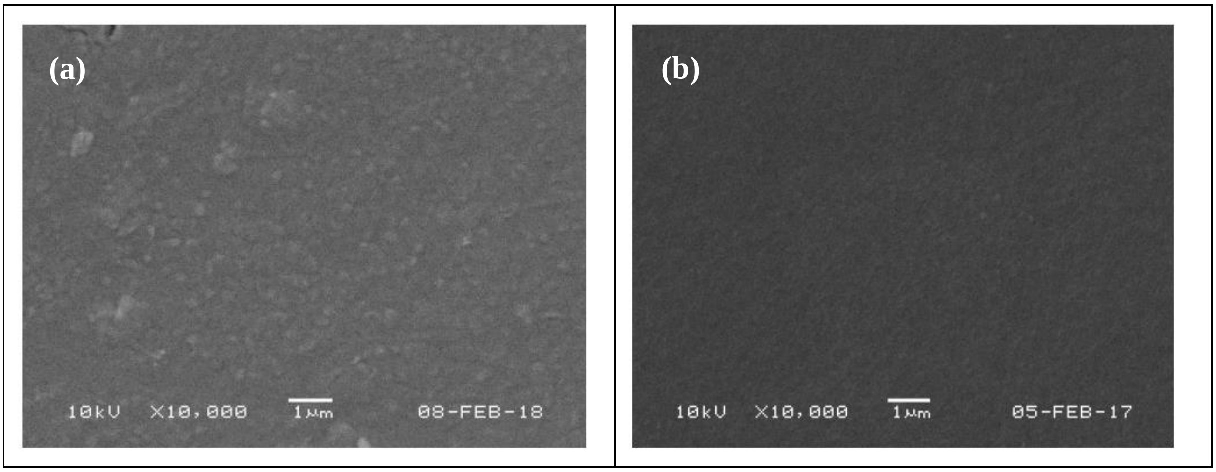


Figure 4-26 SEM image of old membrane (a) compared to a new membrane (b) (Mag. 10.000x)

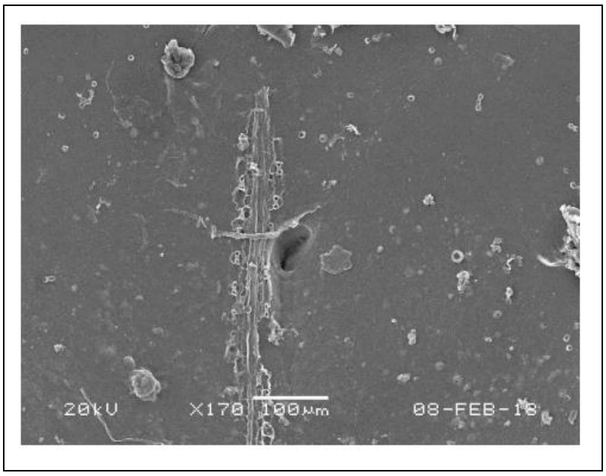


Figure 4-27 Damaged spots in the old membrane (Mag.170x)

The rate of development of irrecoverable fouling of the membrane is an important aspect to consider within the treatment performance as it might determine its economic feasibility. Membranes represent an important percentage of the CAPEX, and therefore their lifespan is critical (EIPWater, 2013). Although this resistance is supposed to be developed over the years, the poor conditions found in the 1 year-old membrane suggest that this factor might require closer attention for this type of wastewater.

Table 4-7 Elemental composition of membrane surface by EDX analysis

Membrane\Element	C	O	N	Ca	F
New membrane	54	4	0	0	42
Old membrane	17	16	38	2	26

4.5 Optimization of membrane operation

4.5.1 Contextualization and preliminary studies

As explained in section 4.4.1, membrane performance during this study and the previous period of operation with synthetic wastewater was not correlated with the one of full-scale systems treating similar wastewater. Particularly very high TMP and low fluxes were obtained and, considering the small particle size measured on sludge; it is expected to be caused by rapid development of cake layer after backwashing. The suction of the permeate pump was identified also as an additional factor contributing to compaction of that layer.

Given Beaubien et al. (1996) observations that critical flux increases by increasing the cross-flow velocity for systems operating at “high” TMP, a positive effect in permeation rate is expected from an increase in cross-flow velocity in this case. Particularly a linear correlation between those factors was found in that study. Instead, other study performed on sludge from a food industry showed a bigger decrease on plateau TMP from 550 to 100 mbar when increasing cross-flow velocity from 1.5 m/s to 2.0 m/s versus a lower decrease from 750 to 550 mbar changing it from 1.0 m/s to 1.5 m/s (Odriozola, 2017). Consequently, it was decided to test the effect of increasing cross-flow velocity on the permeability for the reactor of study.

Considering the previously mentioned observations regarding the effect of permeate pump in the obtained flux, some preliminary tests were run in order to determine the extent of the cross-flow velocity effect under different flux levels. Flux setpoints of 10, 15 and 20 Lmh were selected based on the regular operation parameters of the system. Additionally, a fourth condition without using the permeate pump was included. It should be noted that in the last condition with no permeate pump backwashing was not applied between cycles.

Results of those preliminary tests are shown in Figure 4-28 for the 4 conditions including Flux, TMP and permeate pressure (to show it was strictly correlated to the TMP values). The correlation of flux with cross-flow velocity showed different tendencies for all tested conditions. With high and medium permeate pump suction rate, and no permeate pump, either linear (R^2_{20}=0.967; R^2_{15}=0.965; $R^2_{no\_pump}$=0.993) or exponential correlations (R^2_{20}=0.980; R^2_{15}=0.986; $R^2_{no\_pump}$=0.999) were identified. This results are in accordance with what was shown by Beaubien et al. (1996) for the operation in the ‘high-pressure” zone, where cross-flow directly relates to TMP. A linear relation was found particularly in that study and it was related to the significant contribution of the gel layer to the resistance if the system. However, the operational pressures defined as high for that study were values higher than 1000 mbar while in this case the effect is observed at pressures ranging from 200 to 800 mbar.

Conversely, for the low suction condition, the only increase in flux was observed from 1 to 1.2 m/s, and in the rest of the velocities, it remained almost constant. These results are in accordance with the almost negligible effect of cross-flow velocity proposed by Beaubien et al. (1996) for the low pressure-filtration, as it this case where TMP values ranged from 40 to 60 mbar.

The negative effect of applying a high suction on system performance was verified during these tests. For the operation at a high flux set-point, an increase in cross-flow velocity up to 1.7 m/s was required to see a significant difference on flux, increasing from 10 to 16 with a correspondent increment in TMP from 600 to 800 mbar. On the other hand, a flux of 18 Lmh was obtained using the permeate pump at 3.7 rpm for a cross-flow velocity of 1.5 m/s and a much lower operational TMP of 143 mbar. Additionally, a value of 17 Lmh was registered for the cross-flow velocity of 1.5 m/s at a TMP of 464 mbar. Moreover, for 1.1 and 1.3 m/s of cross-flow velocity, the highest flux of all set of experiments was measured in operation without permeate pump and even with lower TMP than for medium or high flux set-points.

These results confirm the idea that using the permeate pump is the main factor affecting the reliable evaluation of filterability. However, with the current set-up design, the operation without that pump also differs from full-scale conditions affecting the results extrapolation. Particularly, no backwash can be done and also filtration mode is changed from constant flux to constant TMP. If operation without permeate pump is desirable, a system that allows flux control and backwash cycles should be installed as will be discussed in section 4.5.4.

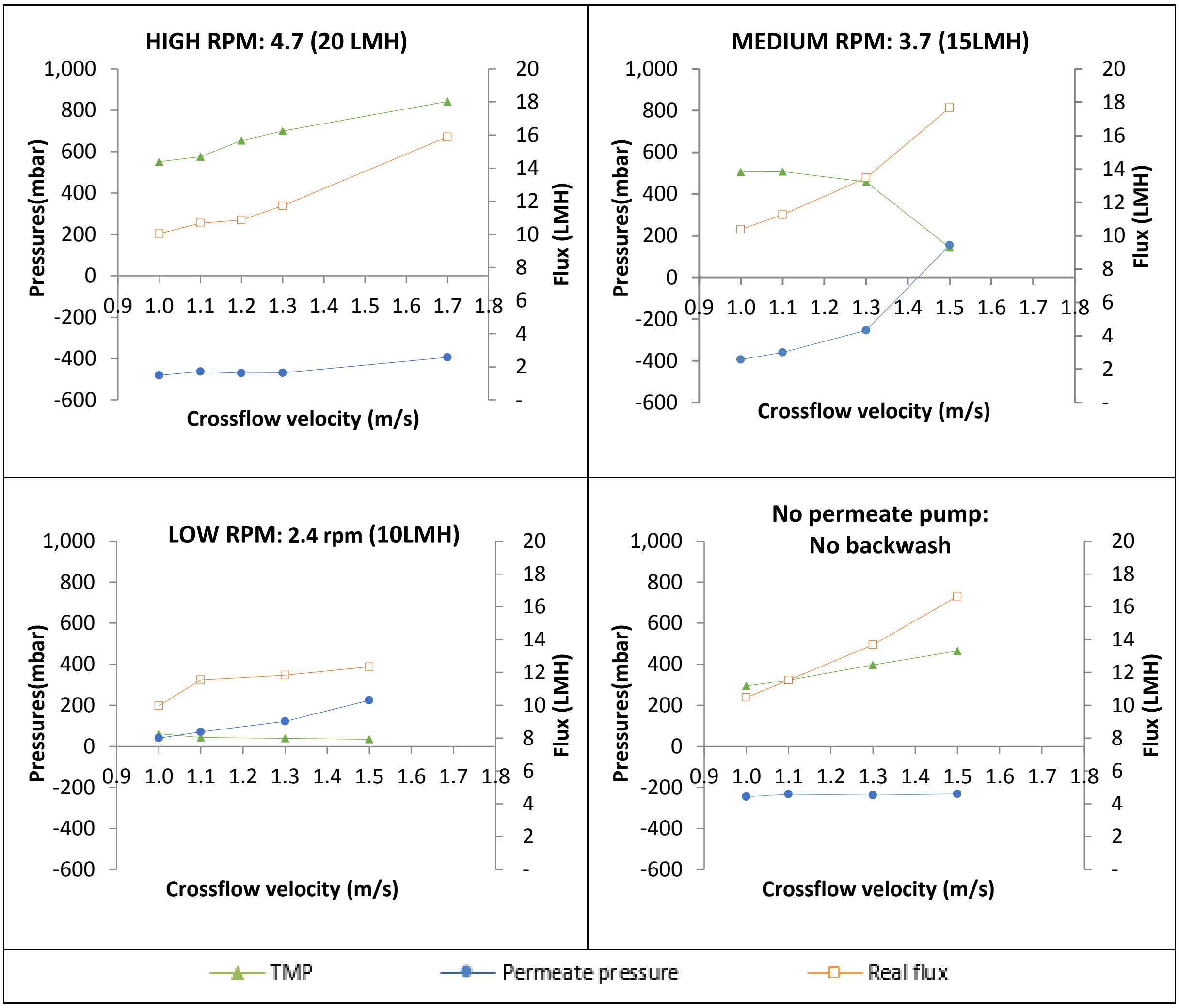


Figure 4-28 Preliminary results of cross-flow velocity effect for 4 different rpms settings

It should be noted that all these experiments were performed in two days and under uncontrolled conditions. Further experiments to estimate in a quantitative and reliable way the effect of increasing cross-flow velocity should be done in order to obtain more decisive conclusions. Considering the presented preliminary results, operation without permeate pump and with manual control of the backwash cycles was selected. Hence, a methodology was proposed for the final tests and the clear definition of such method for it to be applied in other reactors was included as an additional objective of this research.

4.5.2 Complete evaluation of cross-flow velocity and cycle duration effect

The methodology used in these experiments, explained in detail in section 3.2.6, covered a total of 4 different cross-flow velocities and 2 cycle durations. The inclusion of cycle duration as an additional factor for evaluation was done based on the potential possibility of increasing backwash frequency for operation at higher cross-flow velocities. Given the higher shear on the membrane it is expectable a slower cake layer development, and so permeation rate is likely to be maintained for longer without backwashing.

During these trials, sludge and membrane conditions were expected to be constant in order to be able to evaluate only the effect of the desired operational parameters. However, due to the disparity of some conditions of sludge and especially the membrane, 3 experimental stages can be identified. For the first stage, all conditions were randomly distributed during 3 weeks. The second and third stages included the repetition of some especially selected operational conditions to evaluate the effect of variations in membrane and sludge properties.

In full-scale installations, TMP would not increase by an increase in cross-flow velocity as it is governed by the hydraulic head given by the height of the reactor. Conversely, in the laboratory set-up, as observed in the preliminary studies, increasing cross-flow velocity increases significantly the TMP due to a higher feed pressure. Taking that into consideration, in order to resemble full-scale conditions, TMP was maintained at similar values for all the experiments. This was done by increasing membrane pressure for the lower cross-flow velocities through a valve connected to the retentate line. It can be noticed from the box plots of Figure 4-29 that average TMP registered was similar for all conditions. However, higher variability was observed at 1.0 and 1.2 m/s in relation to particular clogging events on the sludge line caused by solids accumulation in the constriction of the valve.

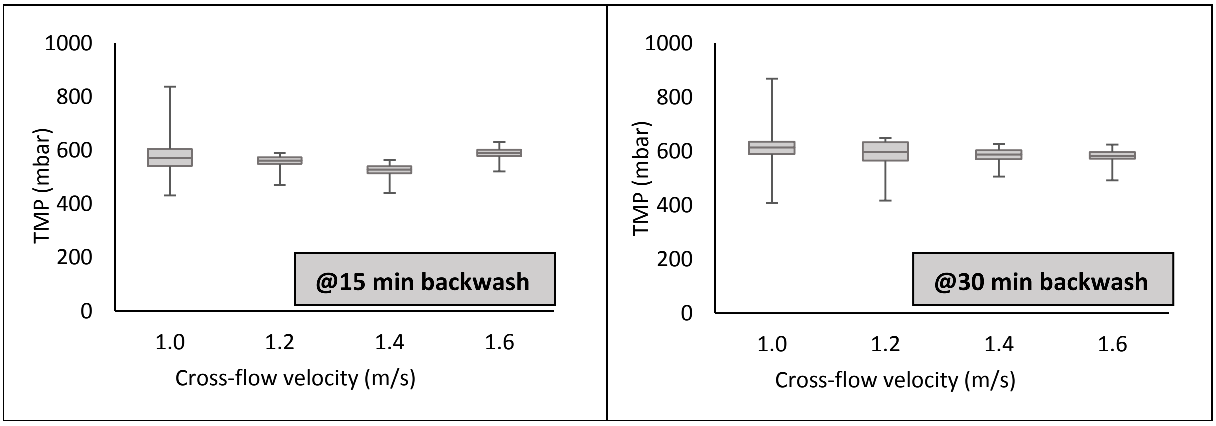


Figure 4-29 TMP value during the first set of experiments at 15 and 30 minutes cycles

Regarding obtained flux during experiments, for all conditions in the 3 sets of experiments, a similar behaviour on real-time flux measurements was observed. As can be distinguished from the two examples shown in Figure 4-30, a rapid decrease followed by stabilization of flux was observed after the first 60-100 seconds of filtration succeeding every backwash sequence. This is most likely related to rapid development of the cake layer during the first seconds of filtration.

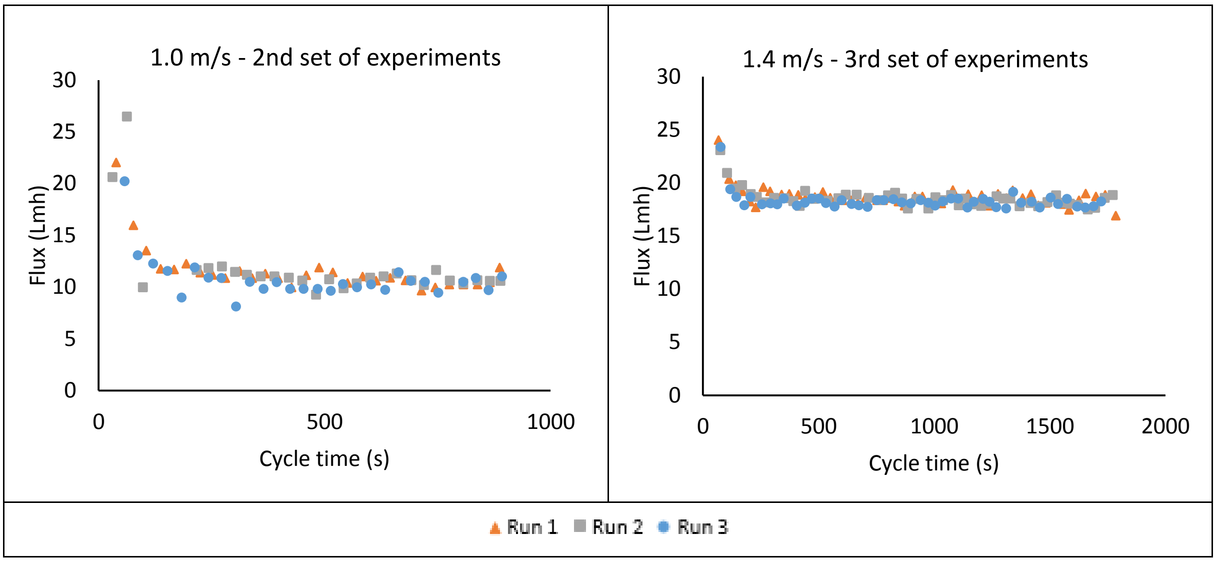


Figure 4-30 Examples of flux-profile during cycles of 15 and 30 minutes

Average values of flux for every condition with the correspondent maximum and minimum considering the 3 runs included in every case are presented in Figure 4-31. The effect of both cross-flow and cycle duration was found significant on the flux results according to the two-way ANOVA analysis (Appendix E). Also interaction of the factors was proven in that test as can be observed from the difference in trend line shape for the two-cycle durations.

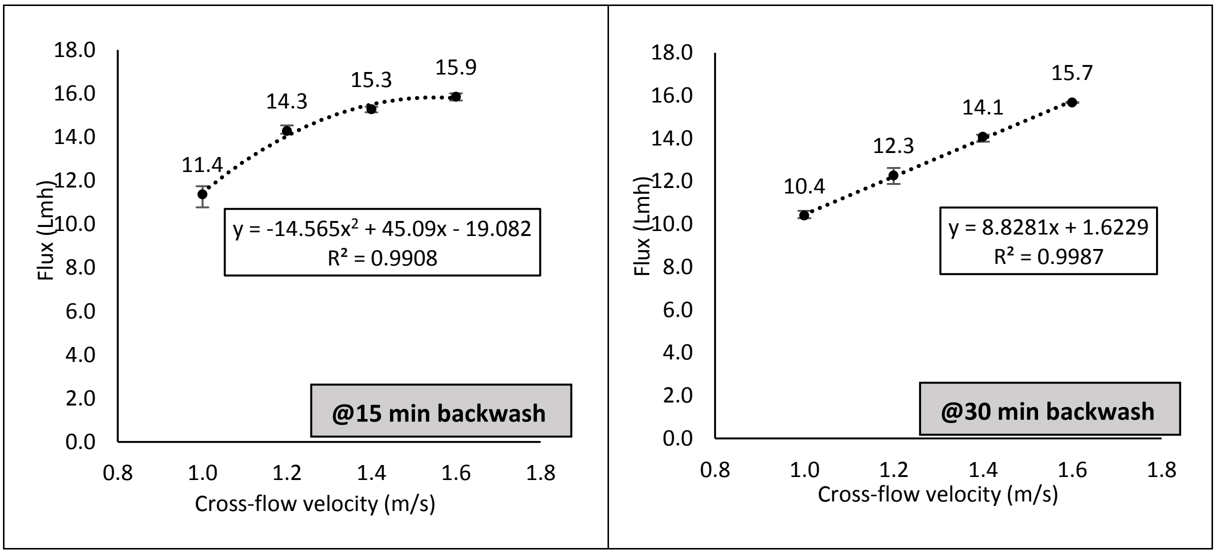


Figure 4-31 Flux measured in the first set of experiments at 15 and 30 minutes cycles

Regarding cross-flow velocity effect, while for the 30 minutes cycle a proper linear correlation was observed (R^2=0.9987), for the operation with more frequent backwash, a polynomial regression was identified to have the best adjustment (R^2_{linear}=0.8767, $R^2_{exponential}$=0.851 and $R^2_{logarit}$=0.9177). Particularly the increase of 26% in flux with a 20% increase in cross-flow velocity (from 1.0 to 1.2 m/s) was the most significant effect found in all this set of experiments (Figure 4-32).

For all cross-flow velocities, decreasing backwash frequency from every 15 to every 30 minutes showed a negative effect on membrane flux (Figure 4-32). However, the difference, in general, was reduced for operation at higher cross-flow velocities, and for 1.6 m/s the flux values resulted in almost the same. It is possible that for higher cross-flows cake layer compaction due to less-frequent backwash is deterred by the high shear of the membrane.

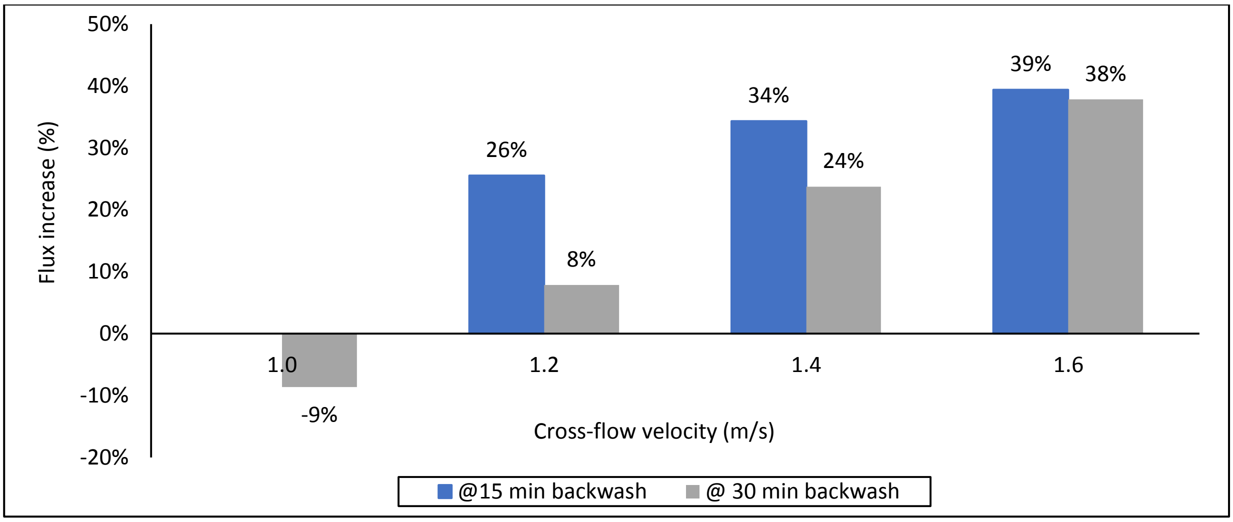


Figure 4-32 Flux difference as a percentage of the value @15 min backwash and 1 m/s cross-flow velocity in the first set of experiments

Table 4-8 summarizes CWP of membrane and sludge filterability properties for every condition. Values marked in bold were found to be outliers using Grubbs test with a confidence level of 95%. Considering that membrane condition deteriorated over time and it can have an influence on the obtained results, the second set of tests for 1, 1.2 and 1.4 m/s at backwash every 15 minutes was done. According to variations on sludge properties also some tests for the longer cycles should have been repeated. However, considering that the new sludge conditions would be more detrimental in relation to the measured fluxes and given the limited amount of time, it was decided not to repeat any test at backwash every 30 minutes.

Table 4-8 Membrane and sludge properties during first set of experiments

Backwash frequency	Cross-flow (m/s)	CWP @30^{o}C (Lmh/Bar)	CST (s)	Viscosity (mPa.s)	d50 (µm)	TSS (g/L)
@15 minutes	1.0	**291**	983	13.985	45.2	12.7
@15 minutes	1.2	115	1255	13.985	45.2	12.7
@15 minutes	1.4	**187**	1119	13.985	45.2	12.7
@15 minutes	1.6	72	1127	14.957	50.5	**14.3**
@30 minutes	1.0	110	900	**10.519**	**35.1**	12.5
@30 minutes	1.2	108	900	**10.519**	**35.1**	12.5
@30 minutes	1.4	68	1100	14.957	50.5	**14.3**
@30 minutes	1.6	69	999	14.957	50.5	**14.3**

During the second set of trials the measured TMPs were similar to the ones in the first set as it is shown in Figure 4-33. From the same figure it can be observed that increasing cross-flow velocity still produced an improvement in the flux but at a lower rate if compared to the first set of experiments. Additionally a linear regression with a good correlation factor between flux and cross flow velocity was obtained in this case in opposition to the first set of experiments.

If the slope of the regression is compared with the one obtained for the cycles of 30 minutes in the first set, cross-flow velocity effect is almost 50% lower in this shorter cycles. However, despite the sludge and membrane conditions in the first experiments at backwash every 30 minutes are more similar to the ones in this second set of measurements, CWP is still lower for the later experiments (Table 4-9), and it might be hindering the effect of cross-flow velocity.

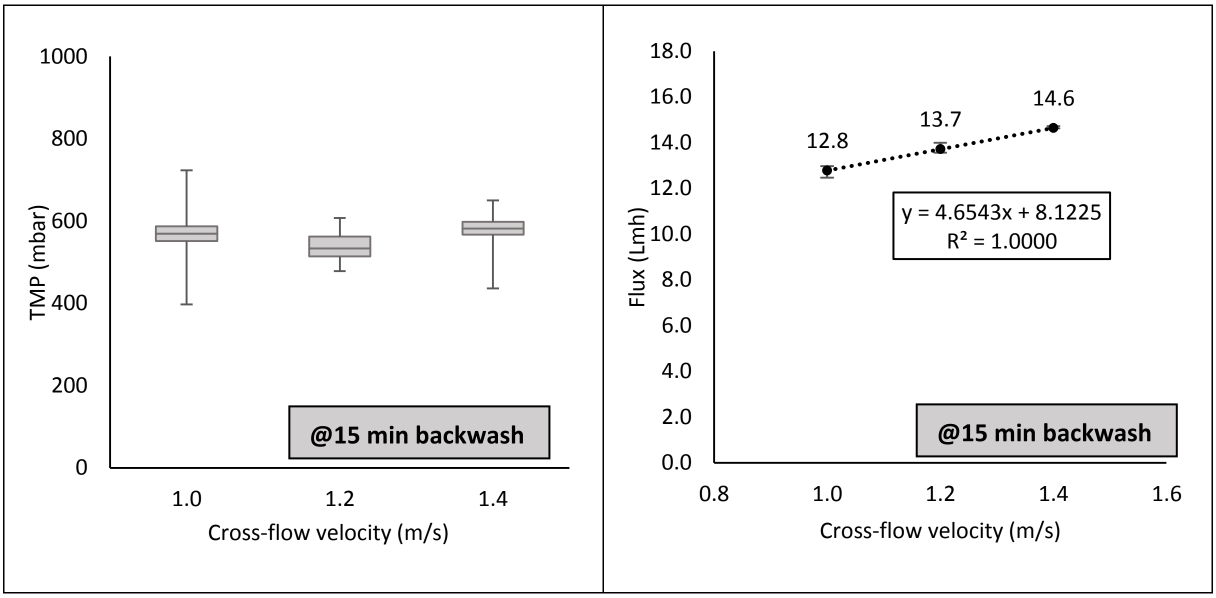


Figure 4-33 TMP and flux during the second set of experiments at 15 minutes cycle

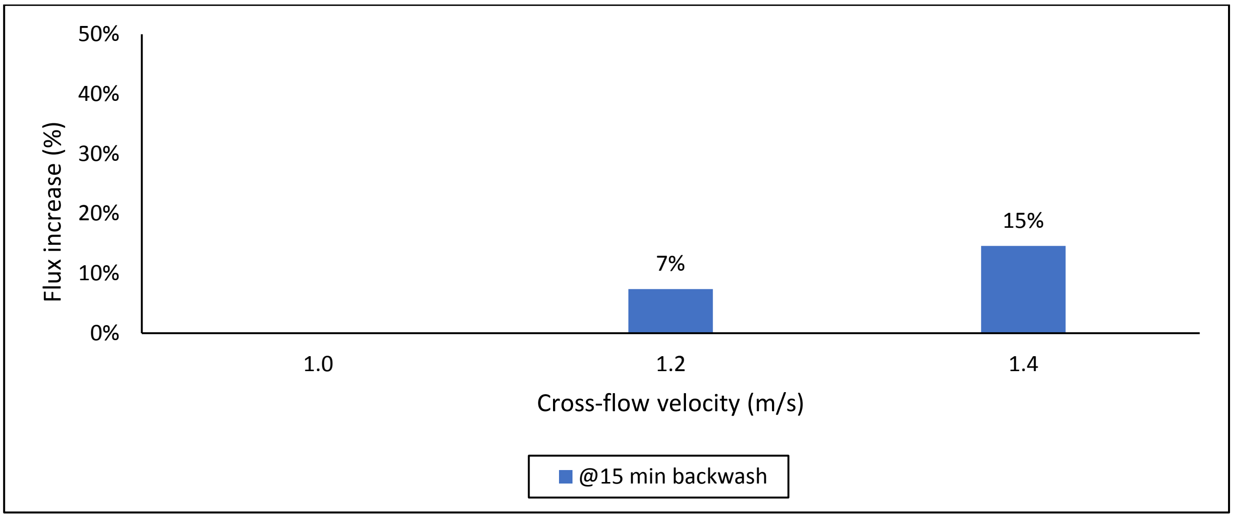


Figure 4-34 Flux difference as percentage of the value @15 min backwash and 1 m/s cross-flow velocity in the second set of experiments

Table 4-9 Membrane and sludge properties during second set of experiments

Backwash frequency	Cross-flow (m/s)	CWP @30°C (Lmh/Bar)	CST (s)	Viscosity (mPa.s)	d50 (µm)	TSS (g/L)
@15 minutes	1.0	58	1293	15.954	56.2	15.0
@15 minutes	1.2	58	1293	15.954	56.2	15.0
@15 minutes	1.4	56	1293	15.954	56.2	15.0

Analysing results of the CWP measured after every day of experiments; it was noticed that the condition of the membrane was decreasing over time reaching very low values for the second set. Consequently, a CIP was run planning to repeat some of the trials in a better membrane condition. By doing so, the effect of the evaluated parameters under different membrane conditions can also be assessed.

The reactor was regularly operated for five days after running the CIP before running the trials in order to reach a steady state of filtration in accordance to Beaubien et al. (1996) procedure. All the experiments were conducted during the last three days of reactor operation included in this research, and the selected conditions were 1, 1.2 and 1.4 m/s for the 15 minutes cycle and 1 and 1.4 m/s for the longer one. Given the short period of evaluation, sludge characteristics were assumed to remain constant and were measured only once (Table 4-10).

The operational values of TMP were adjusted considering the condition of 1.4 m/s cross-flow velocity as it was the highest for this set. However, as it can be noticed from Figure 4-35, higher values were required if compared to the first set of trials with 1.6 m/s. This is in accordance also with the higher feed pressures registered over regular reactor operation for this period. This effect can be related to the increase in viscosity of the sludge.

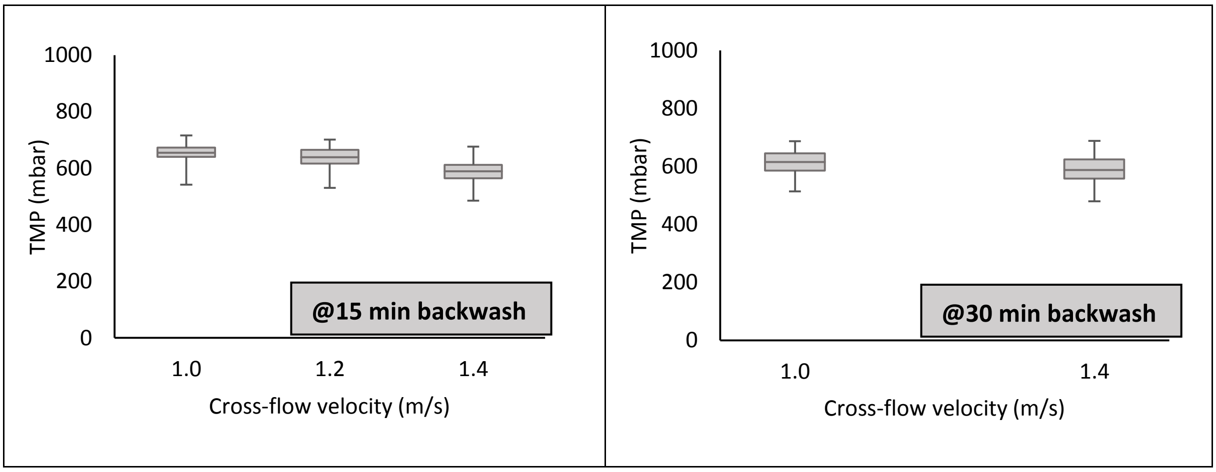


Figure 4-35 TMP during the second set of experiments at 15 and 30 minutes cycles

Regarding flux measurements, in the third set of experiments, all values were higher than its similar conditions within the previous experimental phases, showing the influence of membrane condition in the permeation rate. Yet again the relation of flux with cross-flow velocity resulted in linear for the 15 minutes cycles. Although it is not possible to define a regression for the 30 minutes cycles as only two cross-flow velocities were tested, a slope was calculated for comparison. In opposition to the results of the previous experiments, the effect of increasing cross-flow velocity on measured flux was found higher for the shorter cycles in this set. The increment of flux every 20% increase in cross-flow velocity dropped from 18% to 10% for the case of the 30 minutes cycle whereas for the cycle of 15 minutes it doubled from 7% to 15%.

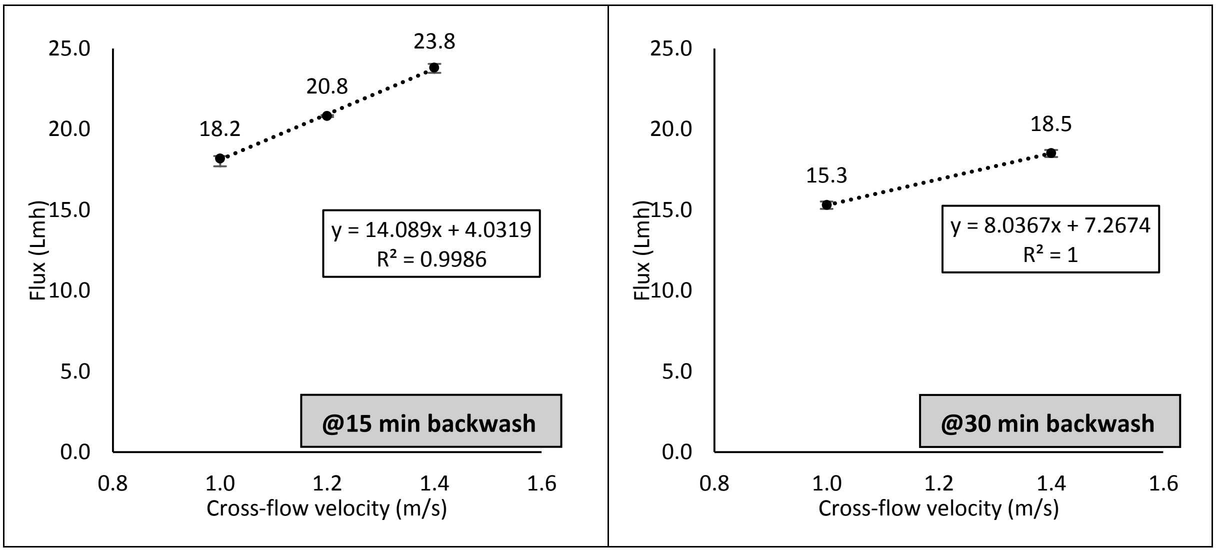


Figure 4-36 Flux during the third set of experiments at 15 and 30 minutes cycles

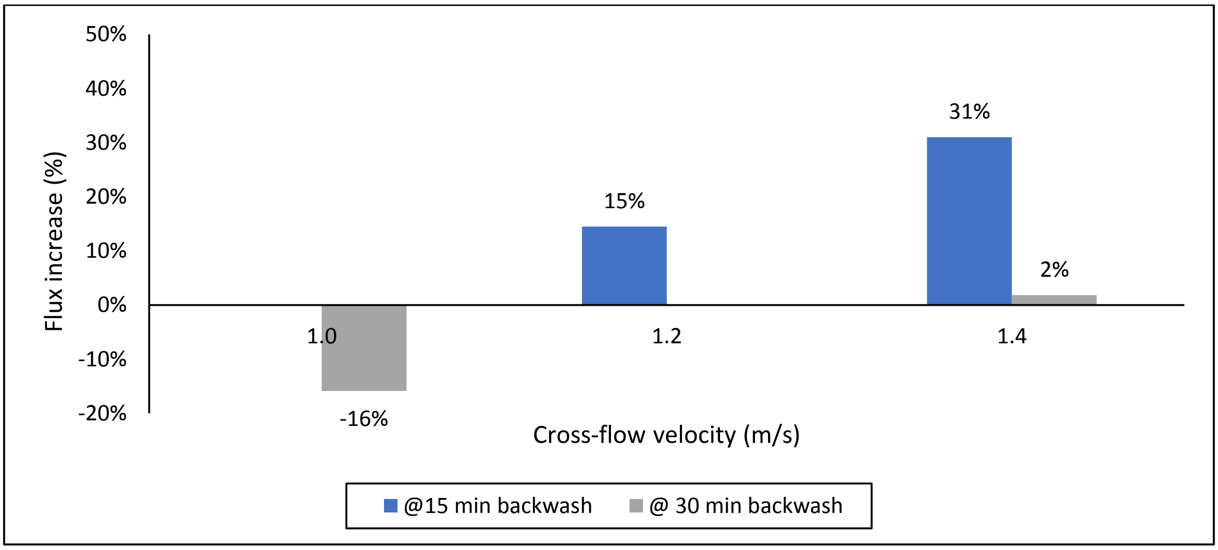


Figure 4-37 Flux difference as percentage of the value @15 min backwash and 1 m/s cross-flow velocity in the third set of experiments

Table 4-10 Membrane and sludge properties during the second set of experiments

Backwash frequency	Cross-flow (m/s)	CWP @30^{o}C (Lmh/Bar)	CST (s)	Viscosity (mPa.s)	d50 (µm)	TSS (g/L)
@15 minutes	1.0	395	1957	16.976	46.6	12.8
@15 minutes	1.2	404				
@15 minutes	1.4					
@30 minutes	1.0	**220**				
@30 minutes	1.4					

Additionally, comparing Figure 4-37 to Figures 4-31 and 4-33, a bigger effect of the cycle duration can be observed. This can be related to the fact that the effect of decreasing backwash frequency is favouring the development of an irremovable cake layer (Yigit et al., 2009). This consequence was more evident in the "clean" membrane than in the "fouled" one as in the second; the irremovable cake layer was already established in advance.

The system was operated from 27/02 to 28/02 at 30 minutes cycles, and the measurements at that cycle duration were done on the 28th. A rapid drop in CWP of the membrane from 395 to 220 Lmh/bar after 20 hours of operation in the longer cycles was measured. Although it can be alleged that still a steady state for filtration could not have been reached, over a similar period of operation in the 15 minutes cycle only produced a reduction of CWP from 404 to 395 Lmh/bar. Considering the short duration of these experiments, long-term effect of cycle duration was not possible to assess, but it should be considered in future studies as might worsen even more the results for the longer cycles to the ones in this study.

Despite the obtained results of filterability improvement due to an increase in cross-flow velocities, the selected cross-flow velocity to apply should finally be based on economic evaluation. According to experience from the company, not much benefit would be seen in the CAPEX in this situation. Investment costs in membrane modules and the skid size will be reduced, but bigger pumps would be needed due to the higher power requirement for the system. Therefore, the main difference will be given by pumping costs that will directly affect the OPEX.

According to the energy estimations done following calculations of Martin et al. (2011), for the system of study, an increase in 20% of cross-flow velocity would lead to an increase in energy consumption per m^3 of treated water of 46% (see Appendix F). Considering the presented calculations, a 70% increase in flux should be produced by the 20% increase in cross-flow velocity to operate at the same energy consumption level while the measured for the system was approximately 15%. As a consequence, only in the case of an exponential effect of cross-flow velocity on permeation rate, it might be worth-it operating at higher cross-flow velocities. These results are in accordance with the sensitivity analysis presented by Shoener et al. (2016), identifying cross-flow velocity and membrane life-span as the most influencing parameters in the net present value of cross-flow configuration AnMBR systems. It is for that reason that current studies of optimization are more focused in other strategies to control fouling in order to be able to operate at even lower cross-flow velocities (< 0.5 m/s) to increase the viability of these systems (Shoener et al., 2016).

4.5.3 Methodology definition

The methodology used for the tests was under development throughout this study. For that reason, some observations during the trials lead to suggestions on how to improve the procedure to make it applicable to another laboratory scale as a standard test using less amount of time. Table 4-11 summarizes these considerations and suggestions for the final procedure. It was assumed that even if the modification of the set-up was done, operation at constant TMP would be used for the trials. The resulting suggested procedure for a general case is presented in Table 4-12.

Table 4-11 Advised methodology for further experiments according to observations in this research

Parameter	Methodology used	Observations	Suggested procedure
Cross-flow velocities selection	Preliminary tests were conducted for different flux set-points and also without permeate pump. Cross-flow velocity was arbitrarily increased during those tests and a change in flux rate was observed for all velocities, particularly for the desired configuration without permeate pump. The selection of values for the formal experiments was based on the ones used during that study.	Starting from 1 m/s as it is the usual value used in the company. The range of values depends on kind of sludge as for example in the study of Odriozola (2017) up to 2 m/s required 3 different values should be sufficient to assess the effect but depends also on the width of the range. In some cases studying lower cross-flow velocities effect might also be of interest.	Run a preliminary test by changing cross-flow on one day in steps of 0.1 m/s from 1 m/s to the desired values (lower or higher) up to levels that show a significant difference in flux register at plateau TMP. Select the maximum and minimium cross-flow velocity to test according to these preliminary results.
Backwash frequency selection	The 15 minutes value was used because it was the regular cycle of the reactor operation. Additionally, 30 minutes cycle was selected to have enough difference in the conditions to note the effect. It was not possible to test values in between due to the available time for the tests.	Backwash frequency can show a short-term effect on membrane CWP so it should be defined as an operational condition at least one day in advance to trials. Considering the general backwash duration for full-scale installations were also ‘idle” time should be added, frequencies lower than 15 minutes would not be practically applicable. For observation of effect a very different value should be selected as the second condition but for optimization closer values as 20 minutes can be tested.	First, run the tests with the regular cycle duration. Decrease backwash frequency progressively towards the minimum value to be tested (in case more than 2 are of interest) and run all experiments for every condition in a maximum of 2 days (max 3 cross-flow velocities). If moving from a lower frequency to a higher, run a CIP before.

Parameter	Methodology used	Observations	Suggested procedure
Number of replicates for every condition to test	Triplicates were desired as is the number generally use in order to identify outliers.	The behaviour of the system during the 3 runs done for every condition as well as average flux results was very similar.	Two replicates are enough under normal testing situations.
Membrane permeability effect	The decision not to make a CIP in the membrane before starting was based on its acceptable CWP (around 400). However, because of the last period of the trials, the value of CWP dropped significantly, and a CIP was run. Tests were repeated for the "clean" membrane condition.	The membrane condition showed to affect the results obtained leading even to under or overestimation of the effect of the factors to be evaluated. Evaluation of CWP before starting trials and after every day is an efficient way of assessing changes in membrane condition, especially when cycle duration is modified.	A CIP should be run on the membrane before running the tests and at least 5 days of operation should be waited before running experiments.
Sludge properties effect	Considering the reactor was operating for almost two months before the experiments, sludge properties were not expected to change much. However, evaluation of them every week or twice a week was done finding significant differences. For the last set of experiments that lasted only 3 days, sludge properties can be considered constant, and this situation is preferred for future evaluation.	Filterability properties of the sludge should be considered. The shorter the duration of all experiments the lower the probability of observing changes in sludge conditions.	Once 3 conditions of cross-flow velocity are selected all the trials for every cycle duration can be run in one or maximum two days, especially considering that two replicates for each give enough information. Assuming all tests can be run in one week, evaluation of sludge characteristics only once would be enough.

Parameter	Methodology used	Observations	Suggested procedure
TMP control during a test	TMP was maintained constant at the highest level measured for the highest cross-flow velocity selected. This was done using a valve connected to the sludge recirculation line to restrict the flow and consequently increase the pressure.	Operating at constant TMP allowed evaluation of permeability by means of flux. Even though regular operation in the new set-up will be at constant flux, for these experiments, it is advisable to keep constant TMP conditions. Otherwise, modifications can be made to use membrane permeability for comparison of the effects.	Select the desired flux setpoint and fixed the valve position for that setpoint to run the rest of the trials. Maintain the real-time measurement of flux to verify its changes due to the operation at constant TMP.
Influence of previous operating cross-flow velocity	For this particular trials, only one different cross-flow velocity to 1 m/s was evaluated every day, so interference of effect was avoided.	Running various different cross-flow velocities in one day is desirable in order to shorten the duration of these analyses. A strategy to avoid the interference of measurements should be defined.	Run one day the cross-flows in increasing order and the following day in decreasing order.

Table 4-12 Suggested methodology for testing cross-flow and backwash frequency effect

Day	Input	Activities	Outputs
1	CWP of membrane	If CWP < 250 Lmh/Bar (@50 Lmh, 1 m/s) >> Run CIP	CIP
6*	Regular cross-flow velocity of operation Expectable cross-flow velocity in the system	Preliminary tests of cross-flow velocity effect changing from 1 m/s (steps of 0.1 m/s or 0.2 m/s) up to 1.5 m/s (higher if necessary to see effect) or also to lower values than 1 m/s if possible. Valve for permeate regulation should be set at a constant position and TMP adjusted to be the same in all trials	Measured flux/TMP for every velocity Selected 3 or 4 levels of cross-flow velocity to test Selected TMP
7	Selected 3 or 4 levels of cross-flow velocity to test Regular cycle duration Selected TMP (position of permeate and feed pressure controlling valves)	Run the system at the selected cross-flow velocities in increasing order (total of 4 cycles for each velocity) and register continuously real flux and TMP for the last two cycles of every condition Run CWP test	Measurement of flux and TMP for the different cross-flow velocities selected CWP of membrane

Day	Input	Activities	Outputs
8	Selected 3 (or 4) levels of cross-flow velocity to test Regular cycle duration Selected TMP (position of permeate and feed pressure controlling valves) Defined next cycle duration to test (20, 30 minutes) if it is of interest	Run the system at the selected cross-flow velocities in decreasing order (total of 4 cycles for each velocity) and register continuously real flux and TMP for the last two cycles of every condition Run CWP test	Measurement of flux and TMP for the different cross-flow velocities selected CWP of membrane System running at the end of the day at new cycle duration
9**	Overnight on cycle duration to be tested Selected 3 or 4 levels of cross-flow velocity to test Selected flux set-point Selected TMP (position of permeate and feed pressure controlling valves)	Run the system at the selected cross-flow velocities in increasing order (total of 4 cycles for each velocity) and register continuously real flux and TMP for the last two cycles of every condition Run CWP test	Measurement of flux and TMP for the different cross-flow velocities selected CWP of membrane
10**	Overnight on cycle duration to be tested Selected 3 or 4 levels of cross-flow velocity to test Selected flux set-point	Run the system at the selected cross-flow velocities in decreasing order (total of 4 cycles for each velocity) and register continuously real flux and TMP for the last two cycles of every condition Run CWP test	Measurement of flux and TMP for the different cross-flow velocities selected CWP of membrane

Day	Input	Activities	Outputs
6 – 10	-	Evaluate sludge characteristics: SS, PSD, CST and viscosity	Sludge filterability characteristics
After finish tests	Flux and TMP profiles for all the different conditions	Data processing: -Plot profiles of flux and TMP -Calculate average flux and permeability for every condition combining results of runs at increasing and decreasing cross-flow values. -Evaluate CWP variation during experiments -Calculate the rate of flux/permeability increase as a function of cross-flow velocity and cycle duration variations -Estimate cost/benefit of modifying parameters	- Effect of cycle duration on CWP of the membrane in short-term - Achievable flux at the selected operational TMP for every cross-flow velocity (permeability) - Optimum cycle duration and cross-flow velocity from an economical point of view based on lab-scale results.

*If no CIP required, it can be day 2. Also can be up to day 7 in case of the weekend.

**Depending on cycle duration and levels of cross-flow velocity can last more than one day

4.5.4 Set-up modifications proposed to resemble full-scale

Considering the observations already explained regarding the effect of using suction to obtain permeate, a modification for the set-up was proposed. Figures 4-38, 4-39 and 4-40 show a scheme of the existent set-up and two alternatives suggested for the new set-up. In the current set-up, flux is controlled by a permeate pump that regulates the TMP by applying suction on the permeate line. The maximum values of TMP that have been reached operating at a set-point of 15 Lmh for the flux (different from the obtained flux of 12 Lmh) were around 600 mbar. It can be noted that height difference from the top of the membrane and permeate tank is approximately 2m and that was the operational pressure in the permeate line (-200-250 mbar) during the tests were not permeate pump was used.

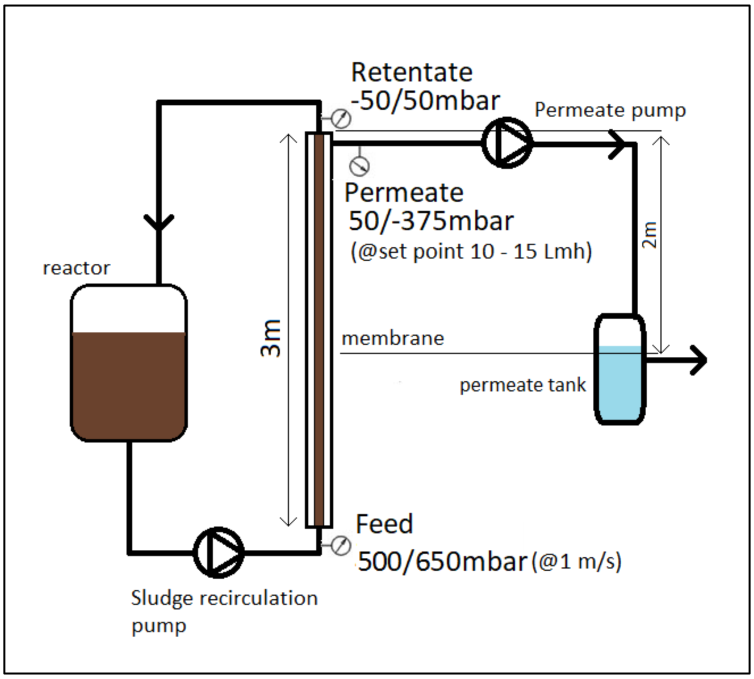


Figure 4-38 Scheme of current membrane set-up and regular operation pressures

The main modification proposed for the new set-up is the replacement of the permeate pump for a controlling valve commanded by a flow meter installed in the line to regulate the permeation rate. In that situation, backwash will be done with an independent pump (same used before for permeation) connected and disconnected through automatic valves commanded by the PLC.

Although the permeation rate is controlled in a similar way to full-scale systems. Still, the maximum pressure achievable on the sludge recirculation line (retentate) is given by the cross-flow velocity (pump pressure) and pressure drop in the membrane. In real systems higher pressures can be reached as up to 6 meters of reactor height is used for the 3 meters long membranes. Therefore, the installation of a valve to increase pressure on the retentate line was proposed in order to emulate pressures of full-scale installations.

However, for the set-up presented as Alternative 1, given the lack of hydraulic head on the retentate side, as it is the case for full-scale installations, the sludge would necessarily suffer a high-pressure drop after the backpressure valve (from 500-1000mbar to 4mbar in the reactor).

The effect on the sludge is similar to the one of hydrodynamic cavitation, a pre-treatment used for sludge digestion. The pressure drop caused by the flow restriction promotes the formation of small bubbles (cavitation) that collapse after the valve causing damage to the cell walls (Lee and Han, 2013). Because of this effect, the filterability properties of the sludge could be significantly affected. Therefore, the specific consequences for the selected operational pressures and sludge should be assessed before final selection of the set-up.

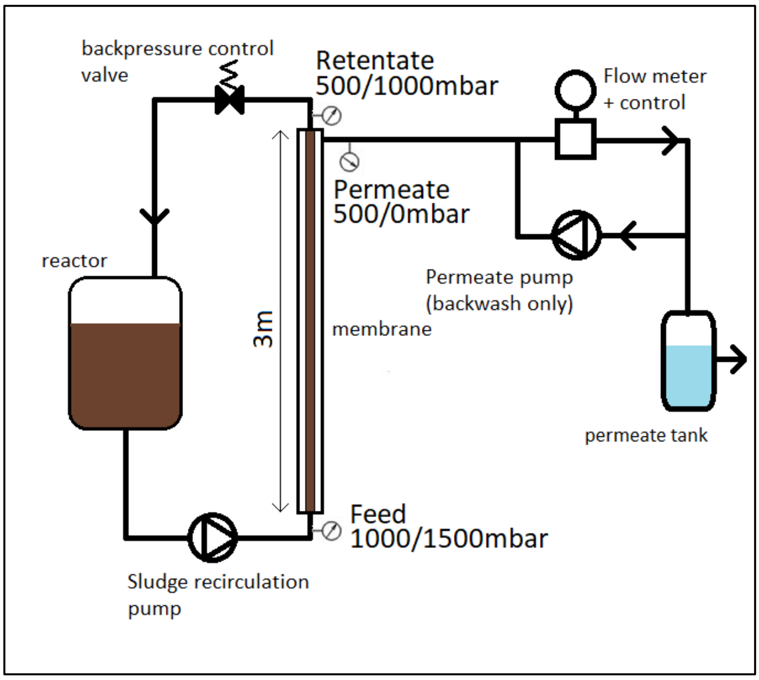


Figure 4-39 Alternative 1 of scheme modification to imitate full-scale pressure in the membrane

The alternative system operation without the backpressure valve and with no permeate pump is presented in Figure 4-40. Considering the range of pressures registered during membrane optimization tests, without restricting the recirculation flow, and using the hydraulic pressure as propelling force for permeation, a maximum TMP of 500 mbar can be achieved in the system at a cross-flow velocity of 1 m/s. Given that full-scale systems generally, operate at TMP lower than that value (100 – 200 mbar), it is expected to be sufficient for membrane evaluation at regular permeation flux.

However, some disadvantages still can be identified for alternative 2. The range of flux is more restricted as maximum achievable TMP is 500 mbar. Additionally, permeate line would still operate at negative pressures (minimum -250mbar). Anyhow, no negative effect of this was observed during the optimization tests operating in those conditions and flux up to 18 Lmh was measured at 1 m/s of cross-flow velocity.

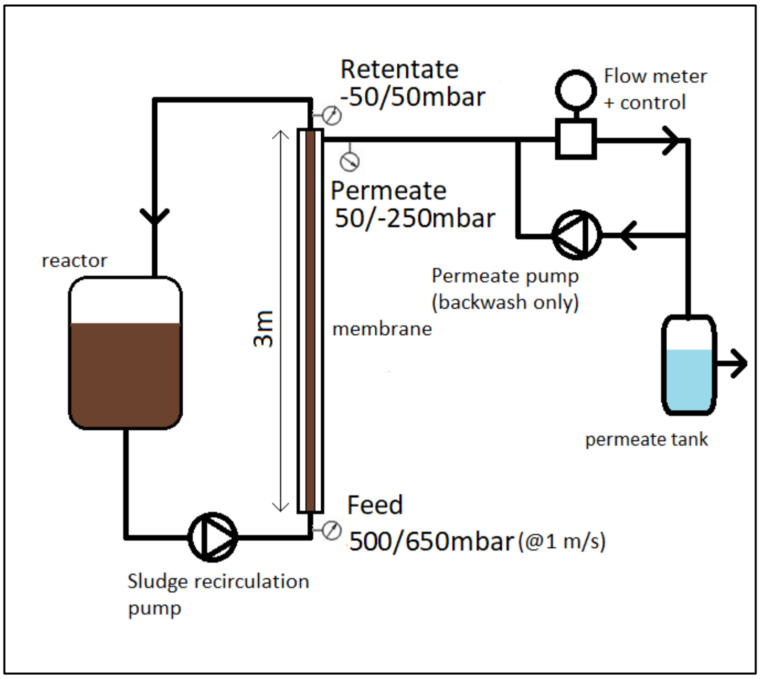


Figure 4-40 Alternative 2 of scheme to avoid a high-pressure drop in the recirculation line

4.6 Overall results

The biological operation of the reactor using real dairy WW was in general satisfactory at the selected VLR of 6 gCOD/Ld which is within the range applied in anaerobic dairy wastewater treatment. The system showed no accumulation of VFAs and was able to deal with some overloading periods due to failures in the control system (up to 12 gCOD/Ld during 12 hours). Additionally, the sludge showed a high SMA compared to other AnMBR. Based on the aforementioned facts it might be possible to test the system efficiency at a higher VLR or at varying VLRs which is of interest due to regular variations of WW production in this industrial sector.

Regarding the effect of intermittent feeding in the biological performance, no significant conclusions can be drawn due to the absence of a control reactor using continuous feeding for comparison. Data from operation in the continuous and intermittent feeding of the same reactor using diluted milk was available for evaluation. However, due to the significant change in the substrate results are not comparable. Nonetheless, the main observed effect of intermittent feeding in that study was in sludge filterability characteristics as no significant differences in biological performance were noticed.

Intermittent feeding effect should be studied further, and it is advisable either to run a parallel reactor in continuous feeding mode or to change the feeding of the same reactor to continuous for comparison. Additionally, based on the observation of the cycle profile, optimization of the duration of the sequence is another improvement that could be done in this system.

The characteristics of the sludge obtained after more than 3 SRTs of operation with dairy WW showed poorer filterability than for the synthetic WW previously used in this reactor. Although MLSS concentrations remain similar, high viscosity and CST values were measured especially after decreasing operational pH from 7 to 6.8.

Contrary to the negative relationship that is expected between average particle size and CST, in this case, an increase in both parameters was observed concurrently. It is expected that despite the presence of bigger particles, the number of small particles in the sludge remained the same as those were reported as the ones controlling the CST(Kim et al., 2002). Additionally, the bigger particle size was linked to the development of filamentous methanogens as it was observed by microscopy of the sludge. Filaments directly affect the dewaterability of sludge and therefore can be a cause for the high CST observed. The slight decrease in the operational pH or the low COD concentrations available during the last hours of the fasting cycles could be the promoters for filament development (Alves et al., 2000).

As far as filtration performance is concerned, values of permeability measured during most of the operation were lower than expected when compared with full-scale results. The maximum achievable average flux was 12 Lmh under regular operation while it is around 20 Lmh in operating industrial systems (Bouman and Heffernan, 2010). This could be related to a poorer sludge quality due to the high shear stress caused by a much higher recirculation rate than in full scale, 180 times a day versus 24 for full-scale. However, similar sludge properties were found in full-scale reactors operating at higher flux values and lower TMP.

For this system, the main cause identified for the unreliability of membrane evaluation was the set-up configuration and particularly, the permeate pump. The high suction on the permeate side generated at high rpm of this pump was found to increase the formation and compaction of the cake layer increasing the TMP and reducing the achievable flux. By removing the permeate pump, values up to 18 Lmh were achieved at the end of the operation for a cross-flow velocity of 1.0 m/s and an acceptable TMP of 400 mbar. Also, much higher permeability conditions were achieved for operation at a lower flux of 13 Lmh using the permeate pump at low rpm mode (2.4 rpm, set-point 10 Lmh) resulting in a TMP of 70 mbar.

From the final experiments of evaluation of the effect of cross-flow velocities, it was observed that the relative benefit of increasing cross-flow velocity varies with the membrane condition. For operation in the regular cycles of 15 minutes, for a low CWP of the membrane, an increase of 0.35% of flux every 1% increase in cross-flow velocity was observed. For a higher CWP condition, this value was doubled to a 0.75% higher flux every 1% increment in cross-flow velocity. The effect of cross-flow velocity at operation in the 30 minutes cycle was lower. An increment of 0.5% in flux every 1% increment in cross-flow was registered during the “clean” membrane condition.

Regarding cycle duration effect, having a backwash cycle every 15 minutes proved to increase the obtainable flux in the system for both low and high CWP of the membrane. The effect was more noticeable in the better membrane condition (from 16 to 20% decrease) as cake layer was not as consolidated as in the “bad” membrane condition (from 3 to 18% decrease).

An estimated evaluation of the effect of increasing cross-flow velocity on the operational costs of the system showed that the increased in flux measured in this study is not enough to compensate the required pumping costs. Operational costs (OPEX) were considered as the most-influential ones assuming the positive effect of reducing membrane modules in CAPEX would be compensated by the additional investment in bigger recirculation pumps. For these reasons, in this particular situation, an increase in cross-flow velocity for regular operation is not recommended as a strategy for fouling mitigation. These results may vary for other systems where the relation of flux and cross-flow velocity might not be linear, as shown in the study of Odriozola (2017).

In addition to the cost evaluation, long-term effect of operation at higher cross-flow velocities should be considered in the selection of the optimal condition. By operating at a higher shear rate, sludge flocs deterioration is expected to affect the filterability. This effect was not possible to assess in this study due to the short-term duration of the experiments. However, the study of Jeison et al. (2009) showed a decrease in critical flux from 35 to 30 Lmh after increasing cross-flow velocity from 1 to 1.5 m/s, presumably due to the decrease in particle size caused by shear stress.

An additional observation from the current study was that for this kind of system regular evaluation of CWP can be used as a good indication of when it is necessary to run a CIP on the membrane. Particularly during the last month of membrane optimization, CWP were performed almost daily, and the values were related to the obtainable flux in the system. An increase of almost 50% was observed during trials for the flux at 1 m/s (without permeate pump) after cleaning the membrane which improved CWP from 56 Lmh/bar to 400 Lmh/bar.

If the period of trials is considered, after one month of operation a sharp decrease in membrane conditions from a CWP of 400 Lmh/bar to 56 Lmh/bar, 15% of the former value was observed. Conversely, the new membrane was operating during more than 40 days before registering the CWP of 400 Lmh/bar. The higher fouling rate for the last operational month can be related to the already mentioned worse filterability properties of sludge. This is an important factor to be considered for the full-scale design as more frequent membrane cleaning decreases the productivity of the system and reduces membrane life-span.

Finally, the proposed modifications on the set-up removing the permeate pump are expected to be beneficial to reach reliable evaluation of filtration performance in laboratory scale reactors. Attaining consisting results of maximum achievable flux is crucial in the evaluation of the applicability of AnMBR technology considering the important share of capital costs that membranes have in this systems.

CHAPTER 5

Conclusions and recommendations

5.1 Conclusions

Dairy wastewater from a cheese producing facility was characterized and treated in a cross-flow AnMBR using a feast-famine strategy. After the operation of the reactor during 107 days, the following conclusions can be drawn:

1. The dairy WW used (mixture) as well as the individual streams (whey and wash water), showed similar characteristics to the ones generally reported for this type of industrial effluent namely: high COD, low pH, low alkalinity and high nutrients concentration. High variations in COD of the whey stream were detected, but the effect on the total COD of the feed was attenuated by the mixture with wash water of lower concentration in a bigger proportion. Although the FOG content measured was lower than for other dairy WW streams, still, the value was around 1.5 g/L (10% of COD) which is higher than normally applied to other high rate anaerobic systems without a pre-treatment step. Therefore benefits of the AnMBRs are still applicable for this WW.
2. Acidification of the wastewater during transport occurred as indicated by the pH drop compared to the values measured in the facility. However, this was not related to the VFAs concentration measured. This is probably because the methodology used for VFAs determination did not allow the quantification of lactic acid which is the main product in lactose fermentation. A greater extent of pre-acidification in the feed tank that was maintained at room temperature for a residence time of approximately 3-4 days was expected. The low pH of the prepared feed was identified most likely as the limiting factor affecting the extent of pre-acidification in this system.
3. The biological performance of the system for the selected feeding regime showed an average COD removal of 98.5% operating at a VLR of 6 g COD/Ld with no VFAs accumulation. Methane conversion measured for this substrate was 0.27 L_{CH4}/kg $COD_{removed,}$ a bit lower than values reported in the literature for dairy WW treatment. However, biogas losses through the mixer of the reactor were identified as a cause, and a value up to 0.30 L_{CH4}/kg$COD_{removed}$ was measured during reduced leakages.
4. The SMA measured on the sludge was 0.6 – 0.8 g COD/g VSSd which is higher than general values found for this type of reactor and within the range of the ones obtained for granular sludge.
5. Operation of the reactor without base addition proved to be satisfactorily reaching a steady state pH of 6.8 without affecting biomass activity or biological efficiency. However, the drop in pH could be related to worse sludge filterability properties measured during the last period of operation as was shown by the calculated correlation.
6. After the feeding was changed from diluted milk to real dairy WW sludge properties related to filterability were affected. By the end of the operation, an increase of CST from values

around 700 s up to 2000 s were measured. The viscosity increased as well from 7-8 mPas up to 16 mPas. This directly affected membrane permeability shown by a decrease in CWP of 85% during the last month of operation reaching values of 56 Lmh/bar. Conversely, a CWP of 400 Lmh/bar was maintained after 40 days of operation of the new membrane installed.

7. The positive expected effect of the feast-famine regime in both biological and membrane performance was not possible to prove as the WW characteristics changed significantly from the data available for continuous operation. Particularly, even poorer sludge quality (higher CST and viscosity) was reached with the real effluent in intermittent feeding than in continuous operation for the synthetic effluent.
8. The positive effect of operation at higher cross-flow velocities in obtainable flux was proved within short-term experiments. The increase rate was significantly different according to membrane condition (measured by CWP). An increase of 1% in cross-flow velocity produced 0.35% higher flux in “bad” membrane condition an up to 0.75% after running a CIP on the membrane.
9. The frequency of backwash also showed a significant effect on the obtained flux and particularly in membrane permeability. Running for 20 hours at 15s of backwash every 30 minutes of filtration reduced CWP of the membrane from 400 Lmh/bar to 220 Lmh/bar. A similar period of operation at the 15 minutes cycle did not have any significant effect in the CWP of the membrane.
10. The best cycle duration from the experiments performed was found to be 15 minutes as reduces membrane fouling and produces a significantly higher flux compared to a 30 minutes cycle. Regarding cross-flow velocity, according to the economic evaluation included in this study, it is not recommended to increase the cross-flow velocity for this particular system. The increase in OPEX is the most relevant factor and in order to compensate for the energy requirements of a 20% higher cross-flow a 70% higher flux should be obtained (only 15% in this case). Additionally, the use of a higher cross-flow velocity can affect sludge filterability in the long-term and therefore, obtained flux increment be even lower than for the present experiments.
11. Through a relatively short experiment, it is possible to evaluate the effect of both cross-flow velocity and cycle duration on the permeability of an AnMBR. This could be useful for the design of full-scale systems as for some particular cases it can be economically favourable to operate at higher or lower cross-flow than the 1 m/s generally used in this company. Also, the optimum cycle duration can be determined in order to operate at the maximum permeation rate with no excessive backwash. This would results in the maximum productivity for the system involving the consequent economic benefit.
12. Regular determination of CWP of the membrane was found to be a good indicator of membrane condition, and therefore, it can be used to define whether a chemical cleaning is required in lab-scale reactors.
13. The existent set-up, based on the suction of the effluent from the membrane through a permeate pump proved not to give reliable membrane performance results compared to full-scale systems. Using the hydraulic pressure difference given by the height difference between the membrane and permeate tank proved to be sufficient to create a TMP around 500 mbar without the negative effect of suction on permeability.

5.2 Recommendations

Considering the results of this research the following recommendations for further studies can be made:

1. Perform a characterization campaign for the wastewater at the treatment plant in order to assess possible variations in effluent COD and pH.
2. Analyse total VFAs of the WW using titration method so as to quantify the lactic acid content to verify the cause of pH drop during transportation.
3. Control of pH on the pre-acidification of feed can be tested at lab-scale to eventually be applied in full-scale systems. It should be considered that this might increase the costs of chemicals for pH control but can also have a positive effect on sludge properties by increasing pH in the reactor and allow higher VLRs in the system.
4. A parallel experiment should be performed using continuous feeding to compare with the feast-famine regime. As this regime might increase the costs of the full-scale installations, due to the necessity of adding a buffer tank, an economic benefit either for operation at higher VLR or Flux should be gained, otherwise continuous feeding would be preferred.
5. Operation at a higher pH through base addition should be tested in order to prove the observed negative effect of lowering the pH on sludge filterability characteristics.
6. Cross-flow velocities and cycle duration are parameters that can be adjusted for an AnMBR depending on sludge properties. To do so, the test developed in this study can be applied to other reactors. Constructing a database with results of these experiments using different wastewater type and sludge characteristics could help in the future to give standard recommendations on optimum operational conditions for every particular reactor.
7. The long-term effect of operating at different cross-flow velocities should be evaluated to define whether the improvement in permeability is maintained or it is affected by the alteration in the shear stress.
8. Regular tests of CWP are recommended to be used to define necessity of chemical cleaning in lab-scale anaerobic reactors. A limit value for this parameter should be set and regular (weekly) evalutations made to follow-up membrane condition.
9. Modification of the set-up to avoid using a pump for permeation is suggested as an improvement. Automatic valves controlled by the PLC are required for the stand-alone operation of the system including backwash cycles. The effect on sludge characteristics of installing a backpressure control valve in the recirculation line should be assessed before modifying the set-up. Also, the range of operational flux obtained for different wastewater and sludge characteristics without that valve should be tested in case it is decided to apply the modification to all the bench-scale set-ups of the company.

References

Abdurahman NH, Rosli YM, Azhari NH (2011) Development of a membrane anaerobic system (MAS) for palm oil mill effluent (POME) treatment. Desalination 266: 208-212

Al-Malack MH, Aldana GR (2016) Performance of anaerobic immersed membrane bioreactor (AnIMBR) treating synthetic dairy wastewater. Desalination and Water Treatment 57: 22200-22211

Alves M, Cavaleiro AJ, Ferreira EC, Amaral AL, Mota M, da Motta M, Vivier H, Pons M-N (2000) Characterisation by image analysis of anaerobic sludge under shock conditions. Water Science and Technology 41: 207-214

Aquino SF, Stuckey DC (2004) The Effect of Organic and Hydraulic Shock Loads on the Production of Soluble Microbial Products in Anaerobic Digesters. Water Environment Research 76: 2628-2636

Beaubien A, Bâty M, Jeannot F, Francoeur E, Manem J (1996) Design and operation of anaerobic membrane bioreactors: development of a filtration testing strategy. Journal of Membrane Science 109: 173-184

Bouman R, Heffernan B (2010) AnMBR, Anaerobic Membrane Bioreactor: From Concept to Full-Scale and Future Outlook Veolia Water Technologies - Biothane Systems International B.V., Tanthofdreef 21, 2623 EW Delft, The Netherlands. http://technomaps.veoliawatertechnologies.com/processes/lib/pdfs/3521-160193_VWT_NL_Poster_IWA_A0_LR_Draf.pdf

Britz T, Van Schalkwyk C, Hung Y (2006) Treatment of dairy processing wastewaters. Waste Treatment on the food processing Industry:1-28.

Brockmann M, Seyfried CF (1996) Sludge activity and cross-flow microfiltration - a non-beneficial relationship. Water science and technology : a journal of the International Association on Water Pollution Research 34: 205

Campos CMM, Luiz FARd, Botelho CG, Damasceno LHS (2004) Avaliação da eficiência do reator UASB tratando efluente de laticínio sob diferentes cargas orgânicas. Ciência e Agrotecnologia 28: 1376-1384

Carvalho F, Prazeres AR, Rivas J (2013) Cheese whey wastewater: Characterization and treatment. Science of The Total Environment 445: 385-396

Cavaleiro AJ, Pereira MA, Alves M (2008) Enhancement of methane production from long chain fatty acid based effluents. Bioresource Technology 99: 4086-4095

Chow L (2001) Solubility of calcium phosphates. In: LC C, Eanes E (eds) Octacalcium Phosphate 94 - 111.

Coelho NM, Rodrigues AA, Arroja LM, Capela IF (2007) Effect of non-feeding period length on the intermittent operation of UASB reactors treating dairy effluents. BIT Biotechnology and Bioengineering 96: 244-249

Couras CS, Louros VL, Gameiro T, Alves N, Silva A, Capela MI, Arroja LM, Nadais H (2015) Anaerobic degradation of dairy wastewater in intermittent UASB reactors: influence of effluent recirculation. environ technol Environmental Technology 36: 2227-2238

Crittenden JC, Trussell RR, Hand DW, Howe KJ, Tchobanoglous G (2012) Appendix C: Physical Properties of WaterMWH's Water Treatment: Principles and Design, Third Edition:1861-1862.

Danalewich JR, Papagiannis TG, Belyea RL, Tumbleson ME, Raskin L (1998) Characterization of dairy waste streams, current treatment practices, and potential for biological nutrient removal. Water Research 32: 3555-3568

Daud MK, Rizvi H, Akram MF, Ali S, Rizwan M, Nafees M, Jin ZS (2018) Review of Upflow Anaerobic Sludge Blanket Reactor Technology: Effect of Different Parameters and Developments for Domestic Wastewater Treatment. Journal of Chemistry 2018: 13

Demirel B, Yenigun O, Onay TT (2005) Anaerobic treatment of dairy wastewaters: a review. Process Biochemistry 40: 2583-2595

Dereli RK, Ersahin ME, Ozgun H, Ozturk I, Jeison D, van der Zee F, van Lier JB (2012) Potentials of anaerobic membrane bioreactors to overcome treatment limitations induced by industrial wastewaters. Bioresource Technology 122: 160-170

Dereli RK, Grelot A, Heffernan B, van der Zee FP, van Lier JB (2014) Implications of changes in solids retention time on long term evolution of sludge filterability in anaerobic membrane bioreactors treating high strength industrial wastewater. Water Research 59: 11-22

Dereli RK, Heffernan B, Grelot A, van der Zee FP, van Lier JB (2015) Influence of high lipid containing wastewater on filtration performance and fouling in AnMBRs operated at different solids retention times. Separation and Purification Technology 139: 43-52

DINAMA, JICA (2010) Informe de Situación sobre Fuentes de Contaminación Difusa en la Cuenca del Río Santa Lucía.

E. Griffin M, McMahon K, Mackie R, Raskin L (1998) Methanogenic population dynamics during start-up of anaerobic digesters treating municipal solid waste and biosolids

EIPWater (2013) Action Group "Anaerobic Membrane Bioreactor for Recovery of Energy and Resources to achieve Sustainable Water Reuse": review of results. https://www.eip-water.eu/action-group-%E2%80%9Canaerobic-membrane-bioreactor-recovery-energy-and-resources-achieve-sustainable-wate-0

Ekama G (2017) A mass balance based steady state model for anaerobic digestion Water research group, Department of Civil Engineering, University of Cape Town.

Fang H, Yu H-Q (2000) Effect of HRT on Mesophilic Acidogenesis of Dairy Wastewater

Fang HHP (2000) Effect of HRT on Mesophilic Acidogenesis of Dairy Wastewater. Journal of Environmental Engineering 126: 1145-1148

Field RW, Wu D, Howell JA, Gupta BB (1995) Critical flux concept for microfiltration fouling. Journal of Membrane Science 100: 259-272

Food and Agriculture Organization of the United Nations (2016) The Global Dairy Sector: Facts

Fraga FA, García HA, Hooijmans CM, Míguez D, Brdjanovic D (2017) Evaluation of a membrane bioreactor on dairy wastewater treatment and reuse in Uruguay. International Biodeterioration & Biodegradation 119: 552-564

Franke-Whittle IH, Walter A, Ebner C, Insam H (2014) Investigation into the effect of high concentrations of volatile fatty acids in anaerobic digestion on methanogenic communities. Waste Management 34: 2080-2089

Gavala H, Kopsinis H, Skiadas I, Stamatelatou K, Lyberatos G (1999) Treatment of Dairy Wastewater Using an Upflow Anaerobic Sludge Blanket Reactor

Ghaly AE (1996) A comparative study of anaerobic digestion of acid cheese whey and dairy manure in a two-stage reactor. Bioresource Technology 58: 61-72

Grethlein HE (1978) Anaerobic Digestion and Membrane Separation of Domestic Wastewater. Journal (Water Pollution Control Federation) 50: 754-763

Gutiérrez JLR, Encina PAGa, Fdz-Polanco F (1991) Anaerobic treatment of cheese-production wastewater using a UASB reactor. BITE</cja:jid> Bioresource Technology 37: 271-276

Hanaki K, Matsuo T, Nagase M (1981) Mechanism of inhibition caused by long-chain fatty acids in anaerobic digestion process. BIT Biotechnology and Bioengineering 23: 1591-1610

He Y, Xu P, Li C, Zhang B (2005) High-concentration food wastewater treatment by an anaerobic membrane bioreactor. Water research 39: 4110-4118

Henze M, Harremoës P (1983) Anaerobic Treatment of Wastewater in Fixed Film Reactors – A Literature Review. Water Science and Technology 15: 1-101

Henze M, van Loosdrecht M, Ekama G, Brdjanovic D (2008) Biological Wastewater Treatment: Principles, Modelling and Design

Ho J, Sung S (2010) Methanogenic activities in anaerobic membrane bioreactors (AnMBR) treating synthetic municipal wastewater. Bioresource Technology 101: 2191-2196

Huang Z, Ong SL, Ng HY (2011) Submerged anaerobic membrane bioreactor for low-strength wastewater treatment: Effect of HRT and SRT on treatment performance and membrane fouling. Water Research 45: 705-713

Hwu CS, Molenaar G, Garthoff J, van Lier JB, Lettinga G (1997) Thermophilic high-rate anaerobic treatment of wastewater containing long-chain fatty acids: impact of reactor hydrodynamics. Biotechnology Letters Biotechnology Letters 19: 447-451

Ince O, Anderson GK, Kasapgil B (1995) Control of organic loading rate using the specific methanogenic activity test during start-up of an anaerobic digestion system. Water Research 29: 349-355

Japan International Cooperation Agency J, Ministry of Housing Land Planning and Environment M (2008) The project on water pollution control and management of water quality in the Santa Lucía river basin. Inception Report, Montevideo, Uruguay. http://mvotma.gub.uy/portal/ciudadania/biblioteca/marco-legal

Japan International Cooperation Agency J, Ministry of Housing Land Planning and Environment M (2011) The project on water pollution control and management of water quality in the Santa Lucía river basin. Final Report, Montevideo, Uruguay. http://mvotma.gub.uy/portal/ciudadania/biblioteca/marco-legal

Jeison D, Telkamp P, van Lier J (2009) Thermophilic Sidestream Anaerobic Membrane Bioreactors: The Shear Rate Dilemma

Jeison D, van Lier JB (2006) Cake layer formation in anaerobic submerged membrane bioreactors (AnSMBR) for wastewater treatment. Journal of Membrane Science 284: 227-236

Jeison D, van Lier JB (2007) Thermophilic treatment of acidified and partially acidified wastewater using an anaerobic submerged MBR: Factors affecting long-term operational flux. Water Research 41: 3868-3879

Jensen PD, Yap SD, Boyle-Gotla A, Janoschka J, Carney C, Pidou M, Batstone DJ (2015) Anaerobic membrane bioreactors enable high rate treatment of slaughterhouse wastewater. Biochemical Engineering Journal 97: 132-141

JICA, MVOTMA (2007) Tthe project on capacity development for water quality management in Montevideo and Metropolitan area. Final report. Volume 3: Supporting report, Montevideo, Uruguay.

Jun D, Kim Y, Hafeznezami S, Yoo K, Hoek EMV, Kim J (2017) Biologically induced mineralization in anaerobic membrane bioreactors: Assessment of membrane scaling mechanisms in a long-term pilot study. Journal of Membrane Science 543: 342-350

Kalyuzhnyi SV, Martinez EP, Martinez JR (1997) Anaerobic treatment of high-strength cheese-whey wastewaters in laboratory and pilot UASB-reactors. Bioresource Technology 60: 59-65

Karadag D, Köroğlu OE, Ozkaya B, Cakmakci M (2015a) A review on anaerobic biofilm reactors for the treatment of dairy industry wastewater. Process Biochemistry 50: 262-271

Karadag D, Koroglu OE, Ozkaya B, Cakmakci M, Heaven S, Banks C, Serna-Maza A (2015b) Anaerobic granular reactors for the treatment of dairy wastewater: A review. International Journal of Dairy Technology 68: 459-470

Kim J-S, Lee C-H, Chang I-S (2001) Effect of pump shear on the performance of a crossflow membrane bioreactor. Water Research 35: 2137-2144

Kim S, Cho S-H, Park H (2002) Effects of particle size distribution on the cake formation in crossflow microfiltration. Water Supply 2: 305

Kimura S (1991) Japan's Aqua Renaissance '90 Project. Water Science and Technology 23: 1573-1582

Kisaalita WS, Pinder KL, Lo KV (1987) Acidogenic fermentation of lactose. BIT Biotechnology and Bioengineering 30: 88-95

Kolarski R, Nyhuis G (1996) The use of sequencing batch reactor technology for the treatment of high-strength dairy processing waste Ann Arbor Press, Inc., Chelsea, MI (United States), United States. http://www.osti.gov/scitech/servlets/purl/395320

Kolev Slavov A (2017) Dairy Wastewaters – General Characteristics and Treatment Possibilities – A Review

Kugelman IJ, McCarty PL (1965) Cation Toxicity and Stimulation in Anaerobic Waste Treatment. Journal (Water Pollution Control Federation) 37: 97-116

Le-Clech P, Chen V, Fane TAG (2006) Fouling in membrane bioreactors used in wastewater treatment. Journal of Membrane Science 284: 17-53

Le Clech P, Jefferson B, Chang IS, Judd SJ (2003) Critical flux determination by the flux-step method in a submerged membrane bioreactor. Journal of Membrane Science 227: 81-93

Leal MCMR, Freire DMG, Cammarota MC, Sant'Anna GL (2006) Effect of enzymatic hydrolysis on anaerobic treatment of dairy wastewater. Process Biochemistry 41: 1173-1178

Lee I, Han J-I (2013) The effects of waste-activated sludge pretreatment using hydrodynamic cavitation for methane production. Ultrasonics Sonochemistry 20: 1450-1455

Lettinga G (2010) The Route of Anaerobic Waste (Water) Treatment toward Global AcceptanceEnvironmental Anaerobic Technology:1-15.

Lettinga G, Field J, van Lier J, Zeeman G, Huishoff Pol LW (1997) Advanced anaerobic wastewater treatment in the near future. Water Science and Technology 35: 5-12

Lettinga G, van Velsen AFM, Hobma SW, de Zeeuw W, Klapwijk A (1980) Use of the upflow sludge blanket (USB) reactor concept for biological wastewater treatment, especially for anaerobic treatment. BIT Biotechnology and Bioengineering 22: 699-734

Liao B-Q, Kraemer JT, Bagley DM (2006) Anaerobic Membrane Bioreactors: Applications and Research Directions. CHIN ChemInform 38: no

Lin H, Peng W, Zhang M, Chen J, Hong H, Zhang Y (2013) A review on anaerobic membrane bioreactors: Applications, membrane fouling and future perspectives. Desalination 314: 169-188

Malamis S, Andreadakis A (2009) Fractionation of proteins and carbohydrates of extracellular polymeric substances in a membrane bioreactor system. Bioresource Technology 100: 3350-3357

Martin I, Pidou M, Soares A, Judd S, Jefferson B (2011) Modelling the energy demands of aerobic and anaerobic membrane bioreactors for wastewater treatment. environ technol Environmental Technology 32: 921-932

McCarty PL (1964) Part Two—Environmental requirements and controlAnaerobic Waste Treatment Fundamentals, 95:123-126.

Meng F, Chae SR, Drews A, Kraume M, Shin HS, Yang F (2009) Recent advances in membrane bioreactors (MBRs): membrane fouling and membrane material. Water research 43: 1489-1512

Meng F, Shi B, Yang F, Zhang H (2007) Effect of hydraulic retention time on membrane fouling and biomass characteristics in submerged membrane bioreactors. Bioprocess and Biosystems Engineering 30: 359-367 DOI 10.1007/s00449-007-0132-1

Meng F, Zhang H, Yang F, Li Y, Xiao J, Zhang X (2006) Effect of filamentous bacteria on membrane fouling in submerged membrane bioreactor. Journal of Membrane Science 272: 161-168

Microtac (2018) Bluewave Microtac. https://www.microtrac.com/bluewave-particle-size-analyzer/

Nadais H, Capela I, Arroja L (2006) Intermittent vs continuous operation of upflow anaerobic sludge bed reactors for dairy wastewater and related microbial changes. Water Science & Technology Water Science & Technology 54: 103

Nadais H, Capela I, Arroja L, Duarte A (2005) Optimum cycle time for intermittent UASB reactors treating dairy wastewater. WR Water Research 39: 1511-1518

Nadais M, Barbosa M, Almeida A, Cunha A, Capela I, Arroja L (2008) Intermittent Operation of UASB Reactors as a Strategy to Enhance Anaerobic Degradation of Dairy Wastewaters

Nadais M, Capela I, Arroja L, Hung Y-T (2010) Anaerobic Treatment of Milk Processing Wastewater

Niejman N (2014) Operator training AnMBR Pentair, The Netherlands.

Novaes RFV (1986) Microbiology of Anaerobic Digestion. Water Science and Technology 18: 1-14

Odriozola M (2017) Report of characterization of sludge from Mars Netherlands - Filterability determination with the Anaerobic Delft Filtration Characterization method (AnDFCm) Delft University of Technology.

ONU (2015) Wastewater Management: A UN-Water Analytical Brief. In: UN-Water (ed), Washington, USA. http://un.org.au/2015/02/03/wastewater-management-a-un-water-analytical-brief/

Passeggi M, López I, Borzacconi L (2012) Modified UASB reactor for dairy industry wastewater: performance indicators and comparison with the traditional approach. Journal of Cleaner Production 26: 90-94

Pavoni JL, Tenney MW, Echelberger WF (1972) Bacterial Exocellular Polymers and Biological Flocculation. Journal (Water Pollution Control Federation) 44: 414-431

Pereira MA, Cavaleiro AJ, Mota M, Alves MM (2003) Accumulation of long chain fatty acids onto anaerobic sludge under steady state and shock loading conditions: effect on acetogenic and methanogenic activity. Water science and technology : a journal of the International Association on Water Pollution Research 48: 33-40

Pereira MA, Pires OC, Mota M, Alves MM (2005) Anaerobic biodegradation of oleic and palmitic acids: Evidence of mass transfer limitations caused by long chain fatty acid accumulation onto the anaerobic sludge. BIT Biotechnology and Bioengineering 92: 15-23

Perle M, Kimchie S, Shelef G (1995) Some biochemical aspects of the anaerobic degradation of dairy wastewater. WR</cja:jid> Water Research 29: 1549-1554

Pollice A, Giordano C, Laera G, Saturno D, Mininni G (2007) Physical characteristics of the sludge in a complete retention membrane bioreactor. Water Research 41: 1832-1840

Pollice A, Laera G, Saturno D, Giordano C (2008) Effects of sludge retention time on the performance of a membrane bioreactor treating municipal sewage. Journal of Membrane Science 317: 65-70

Prazeres AR, Carvalho F, Rivas J (2012) Cheese whey management: A review. Journal of Environmental Management 110: 48-68

Rodgers M, Zhan X-M, Dolan B (2004) Mixing Characteristics and Whey Wastewater Treatment of a Novel Moving Anaerobic Biofilm Reactor. Journal of Environmental Science and Health, Part A 39: 2183-2193

Saddoud A, Hassaïri I, Sayadi S (2007) Anaerobic membrane reactor with phase separation for the treatment of cheese whey. BITE Bioresource Technology 98: 2102-2108

Sato T, Qadir M, Yamamoto S, Endo T, Zahoor A (2013) Global, regional, and country level need for data on wastewater generation, treatment, and use. Agricultural Water Management 130: 1-13

Sethi S, Wiesner MR (1997) Modeling of transient permeate flux in cross-flow membrane filtration incorporating multiple particle transport mechanisms. Journal of Membrane Science 136: 191-205

Shete B, Shinkar NP (2013) Dairy industry wastewater sources, characteristics & its effects on environment

Shoener BD, Zhong C, Greiner AD, O. Khunjar W, Hong P-Y, Guest JS (2016) Design of anaerobic membrane bioreactors for the valorization of dilute organic carbon waste streams. Energy & Environmental Science 9: 1102-1112

Spagni A, Ferraris M, Casu S (2015) Modelling wastewater treatment in a submerged anaerobic membrane bioreactor. Journal of Environmental Science and Health, Part A 50: 325-331

Stuckey DC (2010) Anaerobic Membrane Reactors. In: Fang HHP (ed) Environmental anaerobic technology : applications and new developments:137-161.

Switzenbaum MS (1983) Anaerobic fixed film wastewater treatment. Enzyme and Microbial Technology 5: 242-250

UNEP (2016) A Snapshot of the World's Water Quality :Towards a global assessment. United Nations Environment Programme, Nairobi, Kenya. uneplive.unep.org/media/docs/assessments/unep_wwqa_report_web.pdf

Uruguay_XXI (2015) Informe Sector Lácteo. Uruguay XXI. Promoción de Inversiones y Exportaciones, Montevideo, Uruguay. http://www.uruguayxxi.gub.uy/informacion/wp-content/uploads/sites/9/2016/01/Informe-Sector-Lacteo-Junio-2015.pdf

Van den Broeck R, Krzeminski P, Van Dierdonck J, Gins G, Lousada-Ferreira M, Van Impe JFM, van der Graaf JHJM, Smets IY, van Lier JB (2011) Activated sludge characteristics affecting sludge filterability in municipal and industrial MBRs: Unraveling correlations using multi-component regression analysis. Journal of Membrane Science 378: 330-338

Van Gaelen P (2018) Memthane an update after 4 months KU Leuven University - CREaS, Delft.

van Lier JB, Mahmoud N, Zeeman G (2008) Anaerobic wastewater treatment. In: Henze M, van Loosdrecht M, Ekama G, Brdjanovic D (eds) Biological wastewater treatment : principles, modelling and design:415-456.

Vidal G, Carvalho A, Méndez R, Lema JM (2000) Influence of the content in fats and proteins on the anaerobic biodegradability of dairy wastewaters. Bioresource Technology 74: 231-239

Von Sperling M, de Lemos Chernicharo CA (2005) Biological Wastewater Treatment in Warm Climate Regions IWA Publishing, London

Vourch M, Balannec Ba, Chaufer B, Dorange Gr (2008) Treatment of dairy industry wastewater by reverse osmosis for water reuse. DES Desalination 219: 190-202

Wojdalski J, Dróżdż B, Piechocki J, Gaworski M, Zander Z, Marjanowski J (2013) Determinants of water consumption in the dairy industry

Wu D, Howell JA, Field RW (1999) Critical flux measurement for model colloids. Journal of Membrane Science 152: 89-98

Wu J, Le-Clech P, Stuetz RM, Fane AG, Chen V (2008) Effects of relaxation and backwashing conditions on fouling in membrane bioreactor. Journal of Membrane Science 324: 26-32

Yang K, Yu Y, Hwang S (2003) Selective optimization in thermophilic acidogenesis of cheese-whey wastewater to acetic and butyric acids: partial acidification and methanation. WR</cja:jid> Water Research 37: 2467-2477

Yigit NO, Civelekoglu G, Harman I, Koseoglu H, Kitis M (2009) Effects of various backwash scenarios on membrane fouling in a membrane bioreactor. Desalination 237: 346-356

You HS, Tseng CC, Peng MJ, Chang SH, Chen YC, Peng SH (2005) A novel application of an anaerobic membrane process in wastewater treatment. Water Science and Technology 51: 45-50

Yu HQ, Fang HHP (2001) Inhibition on Acidogenesis of Dairy Wastewater by Zinc and Copper. environ technol Environmental Technology 22: 1459-1465

Zhang J, Padmasiri SI, Fitch M, Norddahl B, Raskin L, Morgenroth E (2007) Influence of cleaning frequency and membrane history on fouling in an anaerobic membrane bioreactor. Desalination 207: 153-166

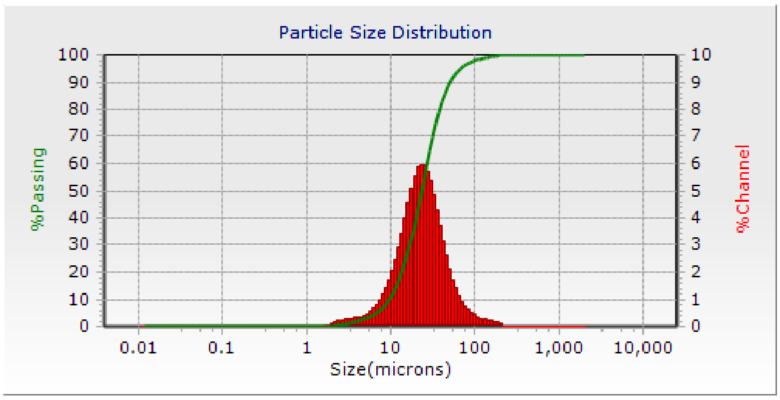
Appendices

Appendix A Selection of flow in PSD measurements

Date: 17/12/17

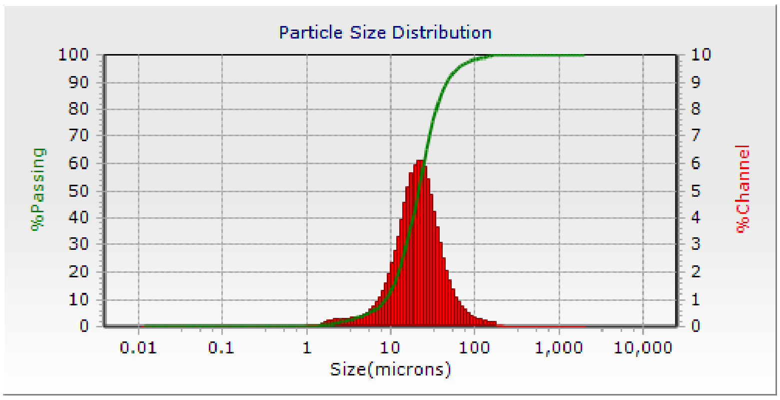
PSD measurement at recirculation flow of 25%

Summary Data	
MV(um):	**28.65**
MN(um):	4.37
MA(um):	17.14
CS:	3.50E-01
SD:	14.76
Mz:	**25.69**
si:	16.75
Ski:	0.37
Kg:	1.348



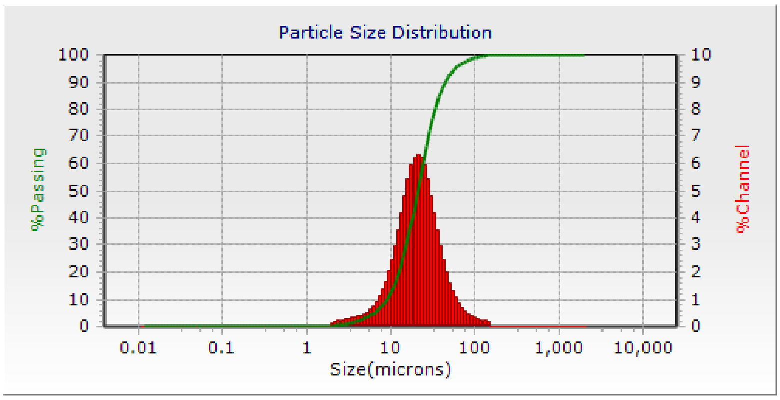
PSD measurement at recirculation flow of 35%

Summary Data	
MV(um):	**25.99**
MN(um):	2.512
MA(um):	14.73
CS:	4.07E-01
SD:	13.33
Mz:	**23.4**
si:	15.24
Ski:	0.355
Kg:	1.376



PSD measurement at recirculation flow of 25% after running at 35% with the same sample

Summary Data	
MV(um):	**25.01**
MN(um):	4.76
MA(um):	15.92
CS:	3.77E-01
SD:	12.62
Mz:	**22.93**
si:	14.17
Ski:	0.353
Kg:	1.326

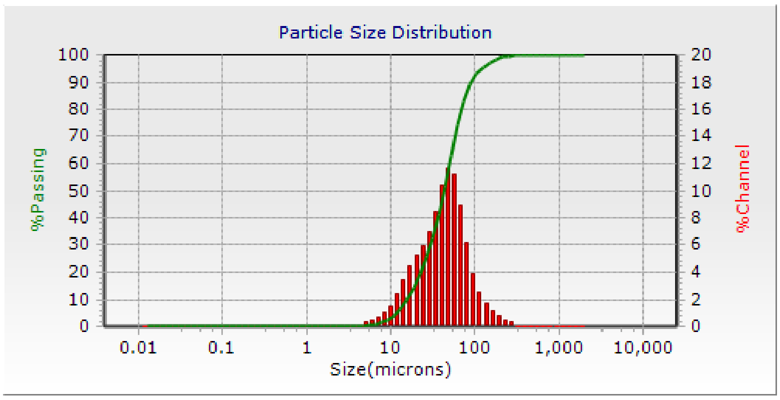


More than 10% reduction in MV and Mz (see bold number) after using a flow of 35%, particle size deterioration at higher flow recirculation.

Date: 09/02/18

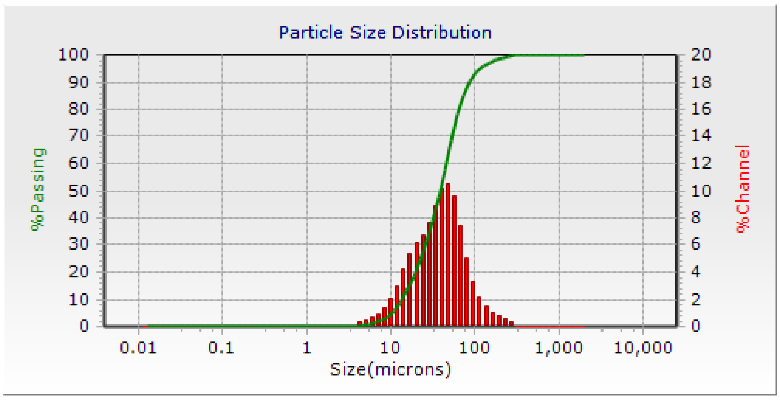
PSD measurement at recirculation flow of 25%

Summary Data	
MV(um):	**50.57**
MN(um):	11.43
MA(um):	31.16
CS:	1.93E-01
SD:	28.14
Mz:	**46.19**
si:	30.31
Ski:	0.2914
Kg:	1.173



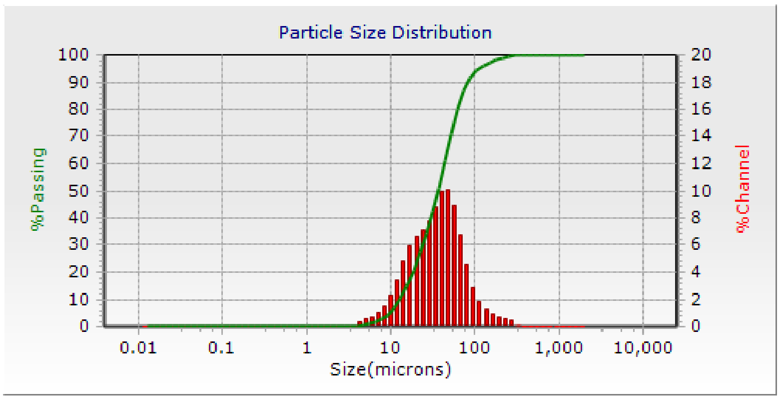
PSD measurement at recirculation flow of 35%

Summary Data	
MV(um):	**46.9**
MN(um):	9.65
MA(um):	27.58
CS:	2.18E-01
SD:	27.03
Mz:	**42.37**
si:	29.34
Ski:	0.336
Kg:	1.175



PSD measurement at recirculation flow of 25% after running at 35% with the same sample

Summary Data	
MV(um):	**45.21**
MN(um):	9.4
MA(um):	26.01
CS:	2.31E-01
SD:	26.18
Mz:	**40.31**
si:	28.59
Ski:	0.356
Kg:	1.178



More than 10% reduction in MV and Mz (see bold number) after using a flow of 35%, particle size deterioration at higher flow recirculation.

Appendix B Additional considerations on cross-flow velocity evaluation methodology

CWP test

- Empty sludge from membrane and return to the reactor.
- Feed water to the membrane on regular flow direction during 10 minutes varying cross-flow velocity from 1 to 1.6 m/s.
- Reverse the flow, empty sludge from reverse flow line and return to the reactor
- Feed water to the membrane on reverse flow direction during 5 minutes varying cross-flow velocity from 1 to 1.6 m/s.
- Return flow to the normal direction.
- Set the desired flux on the computer to 50 Lmh and the cross-flow velocity to 1 m/s. Measure the flux 3 times during 2 minutes and register the average TMP value for the duration of the experiment. Calculate the CWP.
- Stop the recirculation pump and return the system to the normal configuration and set-points of operation.

Backwash verification

- Insert a measurement glass full of water (register volume) below the exit of the permeate line below the permeate tank with the outlet valve closed.
- 2 seconds before backwash starts open outlet valve to allow water for backwash to be sucked out from the glass.
- 2 seconds after backwash finishes, close outlet valve to avoid water to flow from permeate line to the glass.
- A measured volume of water consumed and estimate the backwash flux.

Flux measurements

- Connect the scale to a PC to continuously register the mass of permeate produced during the cycle.
- Open the gas bag to avoid reactor’s low pressure during the experiment due to the volume of permeate lost.
- Register flow vs time (with the scale software) from approximately 15s after backwashing to 15s before backwash. This time is to be able to reconnect the permeate line to the permeate tank during backwash in order to avoid air intrusion.

Schedule

First set of experiments

Backwash frequency	Cross-flow velocity (m/s)			
	1	1.2	1.4	1.6
15 minutes	07/02/18	09/02/18	08/02/18	12/02/18
30 minutes	31/01/18	01/02/18	14/02/18	13/02/18

Second set (repetition of selected conditions)

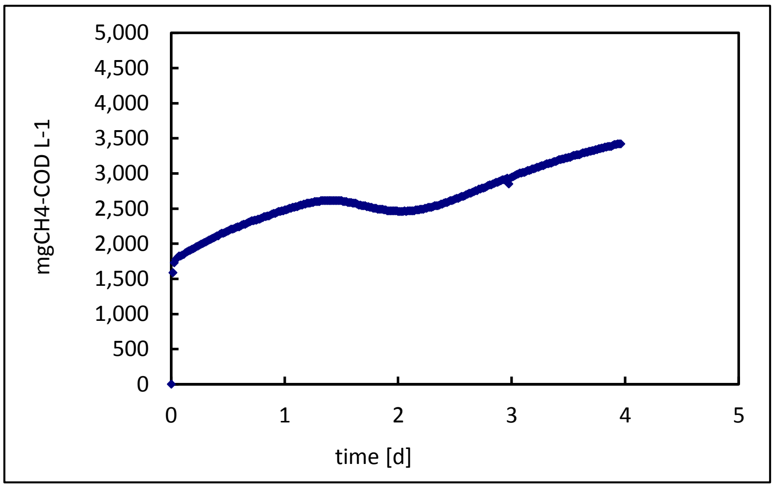
Backwash frequency	Cross-flow velocity (m/s)			
	1	1.2	1.4	1.6
15 minutes	20/02/18	20/02/18	19/02/18	-
30 minutes	-	-	-	-

Third set (after CIP of the membrane)

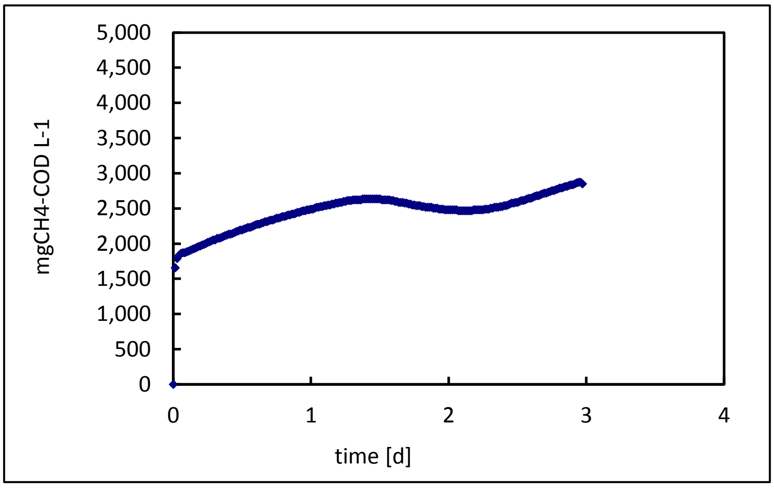
Backwash frequency	Cross-flow velocity (m/s)			
	1	1.2	1.4	1.6
15 minutes	27/02/18	26/02/18	26/02/18	-
30 minutes	28/02/18	-	28/02/18	-

Appendix C **Analysis of residual COD of sludge**

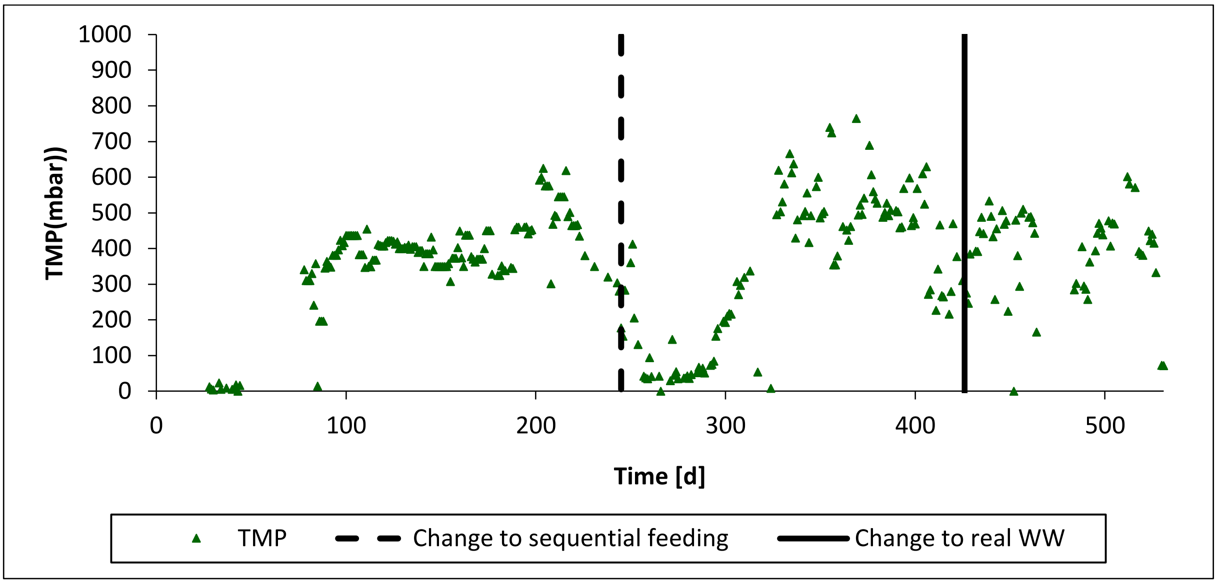
Bottle #	**1**
Vbiomass	50.23
Vliquid	50.0
Vheadspace	240
Conversion factor pressure/volume	0.237
mL CH4 per mg COD	0.397
initial Acetate (mgCOD/L)	*0*
initial VSS (mgVSS/L)	*9,579*

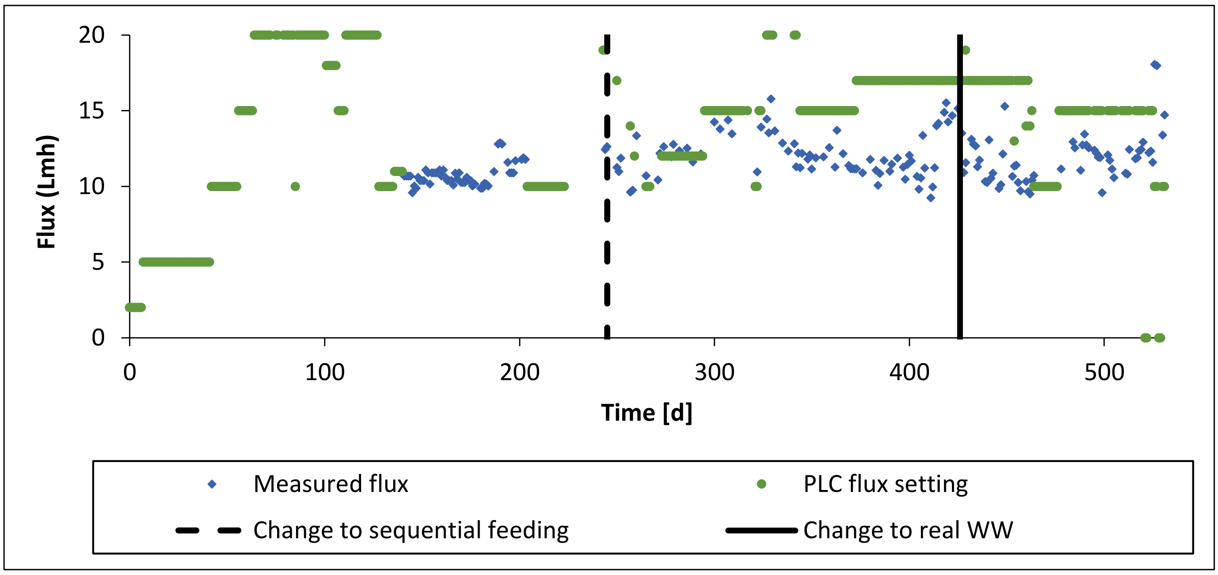


Bottle #	**4**
Vbiomass	50.07
Vliquid	50.0
Vheadspace	240
Conversion factor pressure/volume	0.237
mL CH4 per mg COD	0.397
initial Acetate (mgCOD/L)	*0*
initial VSS (mgVSS/L)	*9,548*



Appendix D **Profile of Flux and TMP during the previous operational period with synthetic WW**





Appendix E ANOVA analysis of cross-flow and cycle duration experiments

First set of experiments

Results

Time	15 min	30 min
1.0 m/s	11.6	10.6
	11.7	10.3
	10.8	10.3
1.2 m/s	14.2	12.3
	14.5	12.6
	14.5	11.9
1.4 m/s	15.1	14.2
	15.4	14.2
	15.3	13.8
1.6 m/s	16	15.7
	15.7	15.7
	15.9	15.6

ANOVA

Anova: Two-Factor With Replication

SUMMARY	15 min	30 min	Total
1.0 m/s			
Count	3	3	6
Sum	34.10747271	31.2	65.30747271
Average	11.36915757	10.4	10.88457878
Variance	0.270412156	0.03	0.40194478
1.2 m/s			
Count	3	3	6
Sum	43.17795691	36.8	79.97795691
Average	14.3926523	12.26666667	13.32965948
Variance	0.042521701	0.123333333	1.422286491
1.4 m/s			
Count	3	3	6
Sum	45.82438107	42.2	88.02438107

Average	15.27479369	14.06666667	14.67073018
Variance	0.017584255	0.053333333	0.466238306
1.6 m/s			
Count	3	3	6
Sum	47.6	47	94.6
Average	15.86666667	15.66666667	15.76666667
Variance	0.023333333	0.003333333	0.022666667
Total			
Count	12	12	
Sum	170.7098107	157.2	
Average	14.22581756	13.1	
Variance	3.33190304	4.267272727	

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Sample	79.63004325	3	26.54334775	376.6006	4.8E-15	3.238872
Columns	7.60479103	1	7.60479103	107.8978	1.61E-08	4.493998
Interaction	2.833187304	3	0.944395768	13.39921	0.000125	3.238872
Within	1.127702889	16	0.070481431			
Total	91.19572447	23				

Second set (repeated) + first set of experiments

Results

Time	15 min	30 min
1.0 m/s	12.9	10.6
	13	10.3
	12.5	10.3
1.2 m/s	13.6	12.3
	14	12.6
	13.6	11.9
1.4 m/s	14.6	14.2
	14.6	14.2
	14.7	13.8
1.6 m/s	16	15.7
	15.7	15.7
	15.9	15.6

ANOVA

Anova: Two-Factor With Replication

SUMMARY	15 min	30 min	Total
1.0 m/s			
Count	3	3	6
Sum	38.4	31.2	69.6
Average	12.8	10.4	11.6
Variance	0.07	0.03	1.768
1.2 m/s			
Count	3	3	6
Sum	41.2	36.8	78
Average	13.73	12.27	13.00
Variance	0.05	0.12	0.72
1.4 m/s			
Count	3	3	6
Sum	43.9	42.2	86.1
Average	14.63	14.07	14.35
Variance	0.00	0.05	0.12
1.6 m/s			
Count	3	3	6
Sum	47.6	47	94.6
Average	15.87	15.67	15.77
Variance	0.02	0.00	0.02

Total		
Count	12	12
Sum	171.10	157.20
Average	14.26	13.10
Variance	1.43	4.27

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Sample	57.55	3	19.18	426.31	1.8E-15	3.24
Columns	8.05	1	8.05	178.90	4.21E-10	4.49
Interaction	4.36	3	1.45	32.28	5.1E-07	3.24
Within	0.72	16	0.04			
Total	70.68	23				

Third set of experiments

Results

Time	15 min	30 min
1.0 m/s	17.9	15.1
	17.7	15.5
	18.3	15.5
1.4 m/s	24.1	18.7
	23.5	18.5
	23.9	18.3

*Results of the run at 1.2 m/s and 15 min were not included as no run at the same cross-flow velocity and 30 minutes was done, so it is not possible to use it for the two-way ANOVA

Anova: Two-Factor With Replication

SUMMARY	15 min	30 min	Total
1.0 m/s			
Count	3	3	6
Sum	53.94013	46.14104	100.0812
Average	17.98004	15.38035	16.6802
Variance	0.108152	0.069834	2.098718
1.4 m/s			
Count	3	3	6
Sum	71.4484	55.5562	127.0046
Average	23.81613	18.51873	21.16743
Variance	0.078856	0.045453	8.468456
Total			
Count	6	6	
Sum	125.3885	101.6972	
Average	20.89809	16.94954	
Variance	10.29279	3.000952	

ANOVA

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Sample	60.40589	1	60.40589	799.2971	2.65E-09	5.317655
Columns	46.77307	1	46.77307	618.9062	7.29E-09	5.317655
Interaction	5.458209	1	5.458209	72.22361	2.82E-05	5.317655
Within	0.60459	8	0.075574			
Total	113.2418	11				

Appendix F Energy requirements estimation for different cross-flow velocities

The following equations (Eq. 1 to Eq. 5) presented by (Martin et al., 2011)were used for estimation of energy consumption for the different cross-flow velocities tested in this study. Some additional assumptions were made to simplify the calculations as the purpose was only to get estimations for comparison and not particular results for design. Additionally also some notions of energy demands provided by Biothane were used in these estimations.

$$E_{tot} = E_{per} + E_{CF,P} \qquad Eq. 1$$

where:

E_{tot} Total energy requirement of the system

E_{per} Energy requirements for permeation

$E_{CF,P}$ Energy requirements for fouling control (cross-flow pumping)

$$E_{per} = Q_P \cdot TMP \qquad Eq. 2$$

where:

E_{per} Energy requirements for permeation

Q_P Permeate flow

TMP Trans-membrane pressure

$$E_{CF,P} = \frac{CFV \cdot S_m \cdot \Delta P}{\xi} \qquad Eq. 3$$

where:

$E_{CF,P}$ Energy requirements for fouling control (cross-flow pumping)

CFV Cross-flow velocity

S_m Cross-sectional area

ΔP Pressure losses

ξ Pump efficiency

$$\Delta P = \frac{4\rho f CFV^2 L}{2D} \qquad Eq. 4\ (Darcy - Weisbach)$$

where:

ΔP Pressure losses

ρ	Fluid density
f	Fanning friction factor
CFV	Cross-flow velocity
L	Length of the membrane module
D	Diameter of the membrane module

$$f^{-0.5} = 4\log_{10}(Re \cdot f^{0.5}) - 0.4 \qquad Eq.5\ (Colebrook's\ relationship)$$

where:

f	Fanning friction factor
Re	Reynolds number

Results from these equations and additional assumptions are presented in the following tables.

Friction energy

Cross-flow (m/s)	Etot (kWh/m3)	Ecfp (kwh/m3)	Eper (kWh/m3)	Qperm. (m3/h)	TMP (kPa)	Flux (Lmh)	Filtration area (m2)	Cross-section area (m2)	ΔP (kPa)
1	1.08	1.07	0.01	0.0008	50.00	17.00	0.05	2.12E-05	27.29
1.2	1.58	1.57	0.01	0.0010	50.00	20.00	0.05	2.12E-05	39.30
1.4	1.90	1.89	0.01	0.0011	50.00	23.00	0.05	2.12E-05	46.60
1.6	2.38	2.36	0.01	0.0013	50.00	26.00	0.05	2.12E-05	57.73

* Procedure of calculation for Etot presented by Martin et al was followed
*Pump efficiency assumed as 65%
*Values of flux used were in accordance with real flux results from the third set of experiments and for 1.6 m/s and estimation following the linear tendency of this experiment was made

ΔP friction

Cross-flow(m/s)	ρ(kg/m3)	Viscosity (Pas)	L(m)	D (m)	f Fanning	ΔP (kPa)
1	1000	0.015	3	0.005195448	0.023631478	27.29
1.2	1000	0.015	3	0.005195448	0.023631478	39.30
1.4	1000	0.015	3	0.005195448	0.020585602	46.60
1.6	1000	0.015	3	0.005195448	0.019528406	57.73

* Density assumed equal to water and viscosity as the average of last measured values

Fanning friction factor

Cross-flow (m/s)	1	1.2	1.4	1.6
ρ(kg/m3)	1000	1000	1000	1000
D (m)	0.0052	0.0052	0.0052	0.0052
Viscosity (Pas)	0.015	0.015	0.015	0.015
Reynolds	346	416	485	554
f fanning	0.0236	0.0219	0.0206	0.0195

*Newtonian fluid behaviour used for estimations despite according to Martin et al. (2011)sludge is Bingham plastic fluid

Final energy estimations

Cross-flow (m/s)	Δptotal est. (kPa)	Etotal est. (kWh/m3)	% increase
1	68.2	2.67	--
1.2	98.2	3.92	47%
1.4	116.5	4.72	77%
1.6	144.3	5.91	121%

* Permeation energy not relevant as it is less than 1% of the recirculation energy
* 2.5*ΔP calculated to include additional losses to friction and safety factor for design