Treatment of Source Separated Greywater Using Microbial Electrolysis Cell and Granular Activated Carbon Biofilter

by

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Abstract

Source-diverted greywater contributes to 60-70 % of conventional wastewater in volume, and has a low organic matter concentration as compared to conventional wastewater. Greywater constitutes a high concentration of surfactants that are toxic in nature for the microbial community, which poses a major obstacle to treat source-separated greywater biologically. This study focuses on the development of an integrated process of anaerobic Microbial Electrolysis Cell (MEC) followed by a passively aerated biofilter system as a polishing step for treating source-separated greywater.

A bench-scale dual-chamber MEC reactor was used to treat greywater. The semi-continuous operation was performed at ambient temperatures over different Hydraulic Retention Time (HRT) of 4 days, 3 days, 2 days and 1 day. An average Chemical Oxyen Demand (COD) removal efficiency of 58.4 %, anionic surfactant removal of 59.7 %, and peak volumetric current density of 0.66 A/m^3 were achieved at an HRT of 4 days. With an HRT of 1 day, the MEC reactor removed 31.7 % COD and 39.7 % anionic surfactants, with a maximum peak current density of 0.65 A/m^3.

MEC reactor produced comparable current density at HRT 1 day to that of HRT 4 days. Hence, MEC effluent from 1-day HRT was treated in a Granular Activated Carbon (GAC) biofilter as a polishing step. The biofilter was operated at HRTs of 30 minutes and 60 minutes. The integrated process provided 99.3% COD removal, and up to 98.7% surfactants removal. Also, the final effluent had no odour or color. The treatability of raw greywater was also assessed with biofilter and the performance was compared to the integrated MEC-GAC biofilter combined system. During the 120 days reactor operation, no process instability/disturbance was observed, and stable performance was achieved throughout. The results are promising in the direction of developing more sustainable treatment of source-separated greywater.

Keywords: Source-separation, greywater, microbial electrolysis cell, biofilter, granular activated carbon

Preface

1. For this manuscript, M. Dhadwal was responsible for experimental design, laboratory experiments, data collection and analysis as well as the manuscript composition. Dr. B.R. Dhar and Dr. Y. Liu planned and supervised the study. All authors contributed to the manuscript preparation.

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Thank you.

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Acronyms

AD	Anaerobic Digestion.
AnBR	Anaerobic Biofilm Reactor.
BOD	Biological Oxygen Demand.
COD	Chemical Oxyen Demand.
CW	Constructed Wetland.
EAB	Electrochemically Active Bacteria.
FOG	Fats, Oil and Grease.
GAC	Granular Activated Carbon.
GW	Greywater.
HLR	Hydraulic Loading Rates.
HRT	Hydraulic Retention Time.
IFAS	Integrated Fixed Film Activated Sludge.
LAS	Linear Alkylbenzene Sulphonates.
MBAS	Methylene Blue Active Substances.
MBBR	Membrane Biofilm Bioreactor.
MBfR	Oxygen-based Membrane Biofilm Reactor.
MBR	Membrane Bioreactor.
MEC	Microbial Electrolysis Cell.
MES	Microbial Electrochemical System.
MFC	Microbial Fuel Cell.
MMBR	Macroporous Membrane Bioreactor.
NTU	Nephlometric Turbidity Unit.
OLR	Organic Loading Rate.

RBC	Rotating Biological Contactor.
RO	Reverse Osmosis.
SBR	Sequencing Batch Reactor.
SHE	Standard Hydrogen Electrode.
SRT	Sludge Retention Time.
TCOD	Total Chemical Oxygen Demand.
TDS	Total Dissolved Solids.
TOC	Total Organic Carbon.
TSS	Total Suspended Solids.
UASB	Upflow Anaerobic Sludge Blanket.
UV	Ultraviolet.
VFA	Volatile Fatty Acids.
VSS	Volatile Suspended Solids.

Chapter 1

Introduction

1.1 Background

Potable water is a resource that is depleting at a higher rate than it is replenished in nature. Climate change, exponential increase in population and wastage are a few of the causes to start with (Brown *et al.* 2013). Many solutions are being put into place to replenish this invaluable resource and counter its shortage. Reuse of treated wastewater is being extensively researched upon and many researchers have devised novel methods to reuse and recycle wastewater and to reduce the usage of potable water (Levine and Asano 2004). Wastewater treatment plants receive feed from sewage, rainwater/surface runoff and greywater. It was a long-standing practice to treat a mixed stream of wastewater and discharge the treated effluent into various receiving environments (wetlands, rivers, lakes, etc.). However, recently it is emphasized that due to the varying characteristics of wastewater (based on source, geography, weather, collection systems), separation at source and applying favorable treatment technologies is a better alternative (Larsen *et al.* 2013). Separate water streams for rainwater (least polluted), greywater (moderately polluted), and blackwater (most polluted) are tested for their characteristics and feasible treatment technologies and methods are developed to treat various streams of wastewater and make the treated effluent re-useable.

The application of MEC systems in anaerobic treatment of wastewater is considered a potentially energy-conserving approach (Escapa *et al.* 2012,Min and Logan 2004). An MEC is an emerging technology that has garnered attention in recent years as it can treat wastewater biologically as well as form value added products (such as H_2, H_2O_2, electric power)(Logan and Rabaey 2012; Rabaey and Verstraete 2005; Rozendal *et al.* 2008). In a typical dual chamber MEC system, there are two chambers (anode and cathode) containing one electrode each, namely anode electrode and cathode electrode separated by an ion exchange membrane. The feedstock is oxidized by the anode respiring bacteria enriched in the anode chamber and released electrons are transferred to anode electrode by extracellular electron transfer mechanism. The transferred electrons then travel via external circuit to the cathode where they combine with protons to evolve hydrogen. To thermodynamically drive the electron transfer in the MEC system a constant potential/voltage is required (Liu and Logan 2004). However, the application of MEC systems for greywater treatment is hardly reported in literature (Couto *et al.* 2015; Hernández Leal *et al.* 2010; Katukiza *et al.* 2014; Khuntia, Hameed, *et al.* 2019; Zhou *et al.* 2020).

Greywater originates from hand basins, showers, laundry, and dishwashers. It does not have a high level of organics that is flushed down the toilets and urinals (Zeeman *et al.* 2008). It can be efficiently treated with treatment methods that require comparatively less energy, such as bio-filters, constructed wetlands, and a combination of physical and chemical treatment systems (Araneda *et al.* 2018; Bolton and Randall 2019; Gulyas *et al.* 2009; Khuntia, Hameed, *et al.* 2019; Wang *et al.* 2019; Zipf *et al.* 2016).

Biofilters are treatment systems that remove contaminants from a feedstock by the action of filtration and microbial activity of the biofilm (Couto *et al.* 2015; Katukiza *et al.* 2014). The major difference between a biofilter and a filtration system is the absence of biological activity in the latter. Interestingly, it has not been reported in literature if re-circulation of wastewater within the system can improve contaminant removal in a bio-filter treatment system.

1.2 Specific objectives

Based on the aforementioned research gaps, the overall objective of this thesis was to evaluate the performance and long-term stability of a MEC system followed by a GAC bio-filter as a polishing step to treat source separated greywater. The performance of the system was assessed based on the non-potable reuse standards for treated greywater re-use as suggested by the Canadian federal guidelines (Drinking Water 2010). First, synthetic greywater was treated with an enriched MEC. Long- term performance (i.e. organic matter removal, anionic surfactant removal and system stability) of the MEC reactor was investigated at different HRTs ranged from 4 days to 1 day. Then, the post-treatment of MEC effluent was performed with a granular activated carbon (GAC) bio-filter at HRTs ranging from 1 hour to 0.5 hours. Moreover, in a separate set of experiments the treatability of synthetic greywater was assessed with GAC bio-filter at HRT ranging from 1 hour to 0.5 hours. It was also investigated if re-circulation of wastewater improves the treatment efficiency in a bio-filter system. The results of the study will provide valuable information and recommendations to further engineer sustainable greywater treatment technologies.

1.3 Thesis organisation

This M.Sc. thesis document the treatment of source separated greywater using MEC and GAC bio-filter at different HRTś ranging from 4 to 1 days in MEC system and treatment of that MEC effluent at HRTś ranging from 0.5 to 1 hour in the GAC-biofilter. The organization of this thesis is as follows.

- Chapter 2 provides a literature review on the current status of treatment studies on source-separated greywater. The review mentions the significance of various treatment methods of greywater separated from domestic wastewater at source. Various biological, chemical, physical and hybrid reactor systems for greywater treatment were presented, compared and discussed in this review.

- Chapter 3 details the reactor configuration, experimental design, analytical methods and calculations throughout the study.

- Chapter 4 presents the results and discussion on the experimental work and data analysis. The greywater treatment performance of the combination of MEC and GAC bio-filter reactors in terms of organic matter removal, anionic surfactants removal, changes in color, odour and system stability was presented and interpreted in this section.

- Chapter 5 summarizes the conclusions from the long-term operation of the reactors. Major performance parameters were specified as references for engineering practice. The recommendations for future work were also proposed in this chapter.

Chapter 2

Literature review

2.1 Source separated greywater

Greywater contributes to 60-70% of conventional wastewater (Gulyas *et al.* 2009), and includes 9–14%, 20–32%, 18–22%, and 29–62% of N, P, K, and organic matter, respectively (Zeeman *et al.* 2008). The characteristics can vary from region to region. The major uses of source-separated greywater are domestic reuse after basic treatment for flushing toilets (Bingley 1996; Christova-Boal *et al.* 1996; Jefferson, Burgess, *et al.* 2001; Nolde 2000; Shrestha *et al.* 2001) for non-agricultural irrigation use, recharging groundwater, and preserving wetlands (Bingley 1996; Christova-Boal *et al.* 1996; Eriksson, Auffarth, *et al.* 2002; Fittschen and Niemczynowicz 1997; Al-Jayyousi 2001; Jefferson, Laine, *et al.* 2001; Nghiem *et al.* 2006; Nolde 2000; Shrestha *et al.* 2001).

The guidelines and standards have become stricter for the reuse of wastewater and demand immaculate effluent quality to qualify for reuse(Exall *et al.* 2006). This has promoted research to develop treatment methods that can deal with a high toxicity of surfactants with a clearer and odour free effluent. Not only that, but the development of a cost-effective method that can be easily deployed and maintained is also a focus for researchers. Many research groups focus on one or more parameters for their research i.e. compliance with reuse standards, cost effectiveness, or near to complete treatment. Reusable/Green

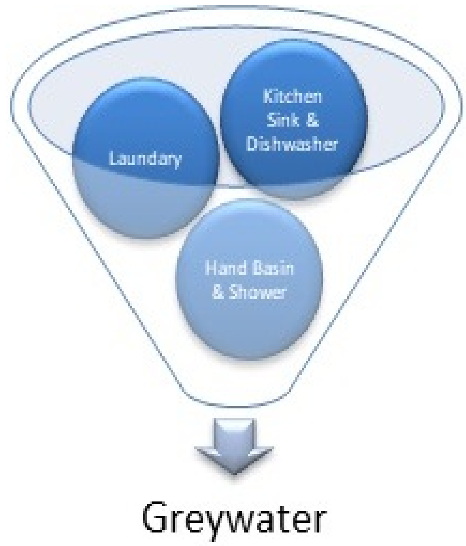

Figure 2.1: Greywater Contributors

energy is also one of the primary focus of the present day's research and industrial sector as fossil fuels are depleting at a higher than ever rate. Efforts are in full swing to recover energy from spent processes or waste and wastewater treatment operations. Technologies such as Upflow Anaerobic Sludge Blanket (UASB), Anaerobic Digestion (AD), Microbial Electrochemical System (MES) have successfully demonstrated that recovering useful products such as chemicals, gases and electricity and consequently energy is possible from simultaneous wastewater treatment operations (Chowdhury *et al.* 2019; Elmitwalli and Otterpohl 2007; M. Gao *et al.* 2018; Liu and Logan 2004). These technologies not only help to facilitate the energy efficient treatment (due to no aeration requirement) but also help to recover energy through them. To achieve effective and efficient treatment of greywater many kinds of aerobic, anaerobic, physical and hybrid treatment technologies have been developed and tested on a laboratory as well as pilot scale and are discussed in the following sections.

2.2 Greywater characteristics

The composition of greywater is highly dependent on its origin namely kitchen, hand basin, bathroom, or laundry greywater (Figure 2.1) as well as the composition of the detergents and chemicals used in the geography of a region(Table 2.2). Typical greywater contains contaminants that are alkaline/acidic substances, Fats, Oil and Grease (FOG), heavy metals, synthetic chemicals and pathogens (Eriksson and Donner 2009; Friedler *et al.* 2005). Boyjoo *et al.* 2013 reported that greywater with high nutrient concentrations is a result of a high fraction of kitchen and laundry effluents in the greywater.

2.2.1 pH of greywater

pH of the greywater is highly dependent on the pH of the incoming water supply and its alkalinity. It is also affected by the characteristics of chemicals/substances used in the household, such as detergents, bleach, fabric softeners, soaps, etc. that can have varying constituents depending upon the country/region of use. Hence, Teh *et al.* 2015 reported a slightly acidic pH of 6.13. On the other hand, Saumya *et al.* 2015 reported basic pH of 11.5 in their study. This variation in pH of greywater can also be credited to the fact that in the above mentioned study they used synthetic greywater for their study. The pH was high due to high alkalinity of the water they used to prepare the greywater. Hence, it is the chosen preparation ingredients of the recipe that is affecting the pH drastically (Saumya *et al.* 2015).

Table 2.1: Greywater Characteristics 1

pH	Alkalinity (mg/L of CaCO)	Chemical Oxyen Demand (COD)	Biological Oxygen Demand (BOD)	Linear Alkyl-benzene Sulphonates (LAS)	Total Suspended Solids (TSS)	Turbidity	Pathogens ((E-Coli/Coliform) (N/100 mL)	Electrical Conductivity (µS/m)	TKj–N (mg/L)	References
7.1-8.3	13.3-28.3	58-294.3	42.1-179.7	14.9-118.3	315-355	133-444	>200.5	1.4-2.9	-	Prathapar *et al.* 2005
-	-	158	59	-	43	33	5.6*105 CFU/100mL	-	-	Friedler *et al.* 2005
-	-	640	-	¬	-	-	-	-	27.2	Elmitwalli and Otterpohl 2007
7.5-7.9	-	25-300	15-140	-	23-50	-	-	-	4.2-20	Lamine *et al.* 2007
-	-	493	-	-	90	-	-	-	21	Lesjean and Gnirss 2006
7.12	-	107-1583	102-215	-	-	-	-	7.52	4.7-47.78	Hernández Leal *et al.* 2010
-	-	681	-	-	-	-	-	-	27.1	Elmitwalli, Shalabi, *et al.* 2007
7.6	-	109	59	0.299	-	29	1.4*105	64.5	15.2	Merz *et al.* 2007
7.12-7.59	454	425-627.5	122-215	-	-	-	-	-	7.9-17.2	Zeeman *et al.* 2008
7-9	-	400-1000	-	1-15	90-200	40-150	-	130-300	-	Ciabattia *et al.* 2009
-	-	724	-	41	-	-	-	-	26.3	Hernández Leal *et al.* 2010
-	-	827-833	-	43.5	-	-	-	-	29.9-41.2	Leal *et al.* 2011

Table 2.2: Greywater Characteristics 2

pH	Alkalinity (mg/L of CaCO)	Chemical Oxyen Demand (COD)	Biological Oxygen Demand (BOD)	Linear Alkyl-benzene Sulphonates (LAS)	Total Suspended Solids (TSS)	Turbidity	Pathogens ((E-Coli/Coliform) (N/100 mL)	Electrical Conductivity (µS/m)	TKj–N (mg/L)	References
6.23	-	146.05	55.6	-	154.63	-	-	-	-	Pathan *et al.* 2011
6.9-7.4	-	179-525	72-182	-	28-146	39-254	-	-	2-13	Abdel-Kader 2013
11.5	76.1	579	290	-	13.3	161.3	-	-	-	Saumya *et al.* 2015
7.2	-	2861	1125	-	996	-	6.9*107	209.7	58.5	Katukiza *et al.* 2014
7.6	-	170	93	-	76	40.4	-	-	-	Couto *et al.* 2015
5.6	25.9	1710	-	163.6	80	-	-	-	32.4	Braga and Varesche 2014
6.13	-	445	349	-	81	-	1.1*108	-	-	Teh *et al.* 2015
-	-	246.63	44.37	18.65	87.3	40.23	-	-	2.81	Chrispim and Nolasco 2017
7.7	-	145	56	8.3	-	35.8	1.8*105	-	-	Zipf *et al.* 2016
7.1	-	477.8	-	-	95.9	15.4	-	277.8	-	Araneda *et al.* 2018
9.76	-	1325	133	-	-	242	-	-	-	Kee *et al.* 2018
-	-	205	-	22	-	-	-	-	-	Wang *et al.* 2019

2.2.2 Electrical conductivity of greywater

Due to the presence of detergents having various salts in them, the greywater usually has good conductivity (Chrispim and Nolasco 2017). Also, conductivity depends on the source of water supply as well. With groundwater having high dissolved contents its conductivity is high. Prathapar *et al.* 2005 recorded electrical conductivity of greywater at 1.4 µS/m. On the other hand, a conductivity of 300 µS/m was recorded by Ciabattia *et al.* 2009 in their study. The type and quality of plumbing used for water supply is also responsible for altering water conductivity. Hence, researchers have reported varying conductivity values ranged from 7.52 µS/m to 277.8 µS/m. (Araneda *et al.* 2018; Hernández Leal *et al.* 2010; Merz *et al.* 2007).

2.2.3 Solids in greywater

In general, greywater has very low content of suspended solids in it. Lamine *et al.* 2007 reported a minimum TSS of only 23 mg/L. Friedler *et al.* 2005 also reported the average TSS of raw greywater in the range of 30-50 mg/L. The main source of these suspended particles are commercial cleaning products, food waste from the kitchen and hair particles from the sinks or showers. However, Prathapar *et al.* 2005 reported TSS values at around 355 mg/L and Katukiza *et al.* 2014 even found TSS to be in the range of 996 mg/L in their greywater stream. This unusual spike in TSS can be explained by the source of greywater being heavily concentrated with detergents from sources such as washing machine and dishwasher. Most of the TSS found in greywater comes from laundry water as the cloths contain sand and silt as well as other suspended particles (Braga and Varesche 2014). Total Dissolved Solids (TDS) is a highly under-reported characteristic of greywater and very few researchers have mentioned TDS values in their reported data.

2.2.4 Biological oxygen demand (BOD) of greywater

A wide range of BOD_5 values (15-1125 mg/L) have been reported in the literature (Table 2.2). This fluctuation can happen due to various factors and mostly indicates to the variability in greywater characteristics with changing habits of the people from where the water is obtained from, in the above-mentioned study a low income group housing facility. The fact that different income group households use different detergents/soaps the greywater coming from low and higher income group houses can have varying characteristics (Gulyas *et al.* 2009, Kujawa-Roeleveld and Zeeman 2006). This also is true for the incoming water supply as higher income group households might have various filters and RO systems to improve the incoming water quality which in turn alters the greywater characteristics coming out of those households.

2.2.5 Chemical oxygen demand (COD) of greywater

A COD value of 1583 mg/L in greywater was reported by Hernández Leal *et al.* 2010 in a study conducted in Netherlands. This abnormally high value came out to be almost double to that of calculated expected value in that geographical region. It was attributed to the time at which sampling was done which favoured a lot of contribution from the laundry discharge for such a high COD in the effluent. This can be validated by the studies done by Katukiza *et al.* 2014; who reported a COD of 2861 mg/L in the discharge from a commercial laundry greywater. However, in a balanced discharge with greywater collected from different sources equally the COD value of 290-850 mg/L (Table 2.2) is obtained which conforms with the calculations done by Hernández Leal *et al.* 2010 for an average value of COD discharged into wastewater stream by a household. The variations are mostly dependent on the source and time of sampling as there may be surge of nutrient/COD discharge if greywater is only collected at certain times of day that coincide with activities such as washing clothes or bathing etc.

2.2.6 COD to BOD ratio

Jefferson, Palmer, *et al.* 2004 reported that greywater had quite high COD:BOD ratio which served as an area of concern for the viability of biological processes in its treatment. A ratio of 2.9-3.6 was reported by them which was almost 2 times to that of conventional wastewater. As per the literature discussed in Table 2.2 some of the studies have reported quite high COD:BOD ratios of upto 9.6 & 7.6 (Kee *et al.* 2018; Kujawa-Roeleveld and Zeeman 2006). On the other hand, Prathapar *et al.* 2005 reported a COD:BOD ratio of only 1.63. Except a few of these exceptions the ratio is almost nearly 2.0 - 3.0 for most of the studies mentioned in the table which suggest that it's best to use biological processes to treat greywater.

2.2.7 Surfactants in greywater

Surfactants concentration is generally reported as the concentration of Linear Alkylbenzene Sulphonates (LAS) in the literature (Araneda *et al.* 2018; Braga and Varesche 2014; Chrispim and Nolasco 2017; Kee *et al.* 2018; Leal *et al.* 2011). Although, there are other types of surfactants (non-ionic and cationic) present in greywater, It is due to the fact that anionic surfactants LAS constitute majority of surfactants present in wastewater and are easily detected as they are Methylene Blue Active Substances (MBAS). Braga and Varesche 2014 reported an average 163.6 mg/ L of anionic surfactants in their greywater study which is more than triple to most of the other reported concentrations. On the other hand, a concentration of merely 0.299 mg/ L of LAS was reported by Merz *et al.* 2007 in their study. These extremities are again attributed to the time of sampling as well as the products being used by the sample population. Surfactants slow down the microbial activity during the treatment process as they are toxic in nature and inhibit the hydrolysis process. Anionic surfactants have been reported to have around 71% anaerobic biodegradability, but the process is very slow (Leal *et al.* 2011).

2.3 Greywater treatment

2.3.1 Aerobic treatment

With the start of 21st century, the enormity of potable water scarcity was realised by the scientists. There was a sudden interest in looking for alternative fresh water sources such as desalination of sea water and exploration of deeper and distant ground and surface water. However, soon enough researchers were quick to identify the ill effects of depleting more freshwater resources. The scientists realised that the technologies such as desalination require more polishing to be efficient and cost effective. Hence, it is one of the technologies that is still being studied for improvement and not applied on a large scale.

Friedler *et al.* 2005 emphasised on efficient use of freshwater and the reuse of greywater as alternative source of water. Onsite treatment of greywater was proposed in this research for reuse in toilet flushing. A conventional combined water treatment method with Rotating Biological Contactor (RBC) as the main biological treatment method followed by sedimentation, sand filtration and disinfection were deployed. The effluent quality reported had 75% removal of COD, 96% BOD, 82% TSS and 98% pathogens. Low COD removal was reported as greywater had more of slowly bio-degradable organics (Friedler *et al.* 2005).

Sequencing Batch Reactor (SBR) for treating greywater from shower rooms of an academic hostel achieved an average COD removal of 90% at different HRT's of 0.6 and 2.5 days and a considerable nutrient removal during the experiment. The authors particularly faced challenges in controlling phosphorus removal at high HRT and nitrogen removal at low HRTś (Lamine *et al.* 2007). Membrane Bioreactor (MBR) technology was also tested to treat greywater at extremely low HRT (2 hrs) and Sludge Retention Time (SRT) (4 days). The authors reported 85% COD removal but struggled with inconsistent nitrogen removal (varying 20-80%) pertaining to low nitrification rates (Lesjean and Gnirss 2006). Another study that used MBR technology with greywater sourced from showers of a locker room in a sports and leisure club reported similar effluent standards with COD removal of around 85%, surfactants removal of 97% and 98% of turbidity reduction (Merz *et al.* 2007). The authors could not guarantee 100% removal of faecal coliforms hence, suggested disinfection step after the MBR treatment to ensure zero coliform contamination in the effluent for reuse purposes.

An extensive study with three pilot-scale systems to treat greywater from different cities by submerged-HRT (SM-SBR) technology was done (Kraume *et al.* 2010). A COD removal of 91%, 95% and 91% in the three consequent plants with odourless effluent of turbidity less than 1 Nephlometric Turbidity Unit (NTU) throughout was reported. However, with continuous operation the membrane clogging was seen, and it was required to be cleaned every 4 months. Although, effluent met the non-potable reuse guidelines, the removal of nutrients was the only challenge between meeting the high mandatory values given by various European directives.

A comparison study of biological treatments with SBR as aerobic, UASB as anaerobic and a combined system of UASB+SBR was done(Hernández Leal *et al.* 2010). Their SBR reactor could remove 90% COD. The anaerobic and combined treatments from this study are discussed in the following sections. Pathan *et al.* 2011 used 54 L volume reactor to treat grey water with RBC technology. The system could achieve a marginal COD removal of 60% and a BOD removal of 53% (Pathan *et al.* 2011). Another RBC system achieved improved removal of 93%-96% BOD, 94% TSS as well as 84%-95% removal of nitrogen with an addition of sedimentation basin followed by Ultraviolet (UV) disinfection. The removal of BOD was enhanced with higher BOD loading rate of up to 5g BOD/m^3 d (Abdel-Kader 2013).

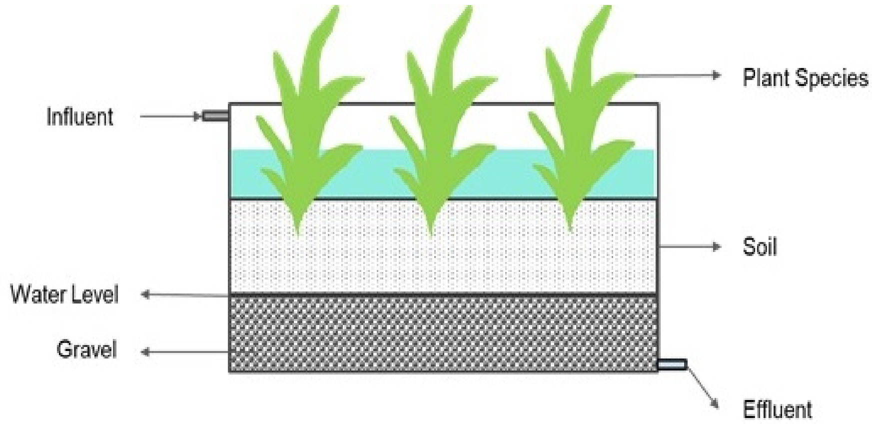

Figure 2.2: Constructed Wetland

Constructed Wetland (CW) (Figure 2.2) is considered to be a sustainable technology that can be used to treat greywater. Saumya *et al.* 2015, studied CW treatment of greywater by rootzone treatment method using Heliconia Augusta plant. A 27% reduction in COD, 48% in BOD, 82% in TSS and 97%in turbidity was reported by them. A sample of 12-15 L was circulated through the wetland setup around the clock for 3 weeks to achieve these effluent standards. A pilot-scale setup was deployed by Chrispim and Nolasco 2017 at a university building in Brazil to collect and treat greywater for non-potable reuse.

The Membrane Biofilm Bioreactor (MBBR) technology-based reactor treated 302 L/d of greywater at 4 hours retention time (Figure 2.3). The system was able to remove COD by 70%, BOD by 59%, TSS by 87% and anionic surfactants by 30% (Chrispim and Nolasco 2017). Eslami *et al.* 2018, then tested nutrient and grey water treatment via the Integrated Fixed Film Activated Sludge (IFAS) process (Figure 2.4). The reactor performance at different Organic Loading Rate (OLR) was studied. Removal of 92.5% COD and 85.24% BOD at 0.44 g COD/L.d of OLR in addition to 89.6% and 86.6% of total nitrogen and total phosphorus removal respectively.

Khuntia, Chandrashekar, *et al.* 2019 built two 50 L Macroporous Membrane Bioreactor (MMBR) and SBR reactors with membrane biofilters made up of commercial grade nylon mesh with 50 and 100 m pores. At an HRT of 1 day the system was able to achieve effluent characteristics with 87% COD removal with the SBR reactor and 91% COD removal with the MMBR reactor. The authors discovered that MMBR achieved steady-state more easily without any complications such as filter clogging. The authors attributed lower COD removal with greywater as compared to sewage water to the presence of higher concentrations of anionic surfactants in the greywater. (Khuntia, Chandrashekar, *et al.* 2019). Zhou *et al.* 2020 operated a bench scale oxygen-based membrane biofilm reactor (O_2-MBfR) to effectively treat organics and nutrients in greywater. The system achieved removal standards of up to 98% of surfactants, 95 % of total COD and 99% of inorganic nitrogen from the waste stream.

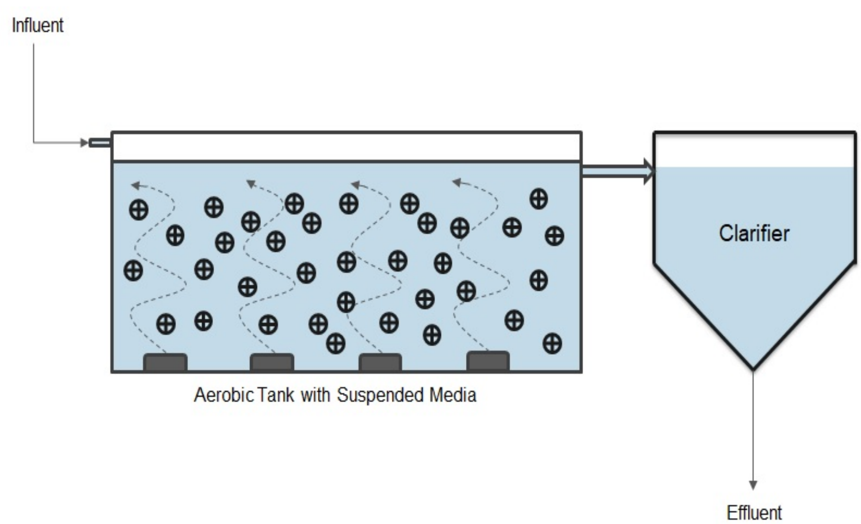

Figure 2.3: Moving Bed Biofilm Reactor (MBBR)

A highly dynamic multi-functional biofilm was achieved by increasing feed loading rates and subsequently lowering the DO concentration from 1.67 to 0.37 mg/ L. The removal mechanism for organics and nutrients were a combination of nitrification and aerobic denitrification in the aerobic region of the biofilm, partial nitrification in aerobic-anoxic biofilm, and partial nitrification and anaerobic denitrification in the aerobic-anoxic-anaerobic region of the biofilm (Figure 2.4). The multidimensional distinct regions were achieved with the presence of diverse functional microorganisms. The study was done on synthetic greywater prepared with International 2011 directions with moderate modifications to adjust nitrogen and pH content of greywater by adding 1.5mL/ L primary sludge and 1.0 M NaOH to the prepared greywater.

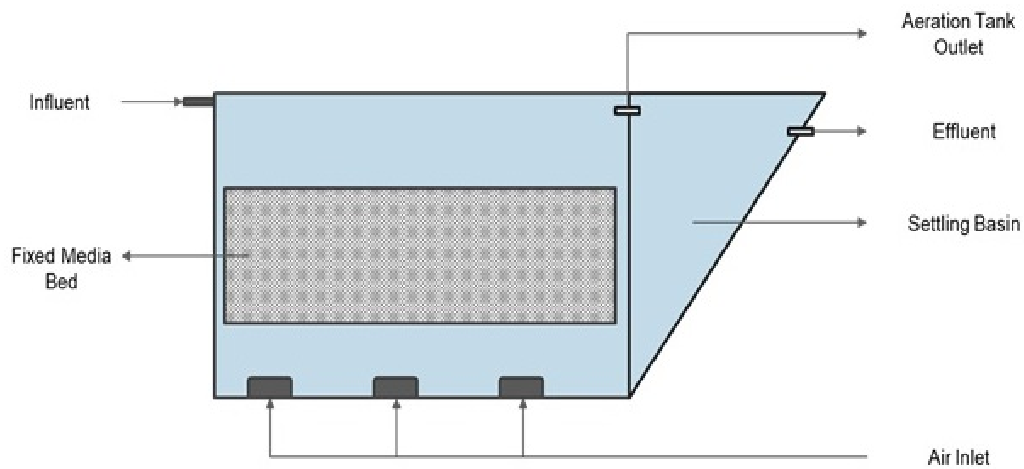

Figure 2.4: Integrated Fixedfilm Activated Sludge (IFAS)

2.3.2 Anaerobic and physical treatment

Anaerobic treatment is particularly helpful in treating the wastewater efficiently and recovering energy from the process of disintegration of organics present in the wastewater. Elmitwalli and Otterpohl 2007 reflected on the anaerobic biodegradability of greywater (Figure 2.5). A 7 L UASB reactor's performance at different HRT to treat greywater at laboratory-scale was studied. A maximum of 64% COD removal with a 764 % anaerobic biodegradability of greywater at an HRT of 16 hours was reported. Interestingly, the authors were the first to compare lower removal of COD with greywater as compared to sewage and attributed the same to the presence of surfactants and high concentration of colloidal COD (Elmitwalli and Otterpohl 2007).

In the following year Elmitwalli, Shalabi, *et al.* 2007, did another study on greywater treatment using UASB technology and could improve COD removal to up to 79% with the same reactor volume as the previous one but at different HRT's and temperatures. Conversion of around 63% of the removed COD to methane at an HRT of 12 hours was also achieved. It was found out that septic tank was not a good pre-treatment method for greywater as prevalent in those days as it removed marginal amount of COD only (Elmitwalli, Shalabi, *et al.* 2007). Hernández Leal *et al.* 2010 performed another study to determine anaerobic biodegradability of greywater. The authors indicated 705% of COD in greywater is biodegradable by anaerobic methods. However, The authors highlighted poor bio-degradability potential of anionic surfactants of around only 3513%. The bio-degradation process with greywater was very slow pertaining to a low hydrolysis constant of 0.02± 0.01d^{-1} (Hernández Leal *et al.* 2010). The authors performed a comparative study on greywater treatment (Section 2.3.2). Their 5L UASB reactor had achieved only about 51% COD removal at an HRT of 12-13 hours. It was observed that due to the poor entrapment/flocculation of COD in the sludge bed the COD removal was low.

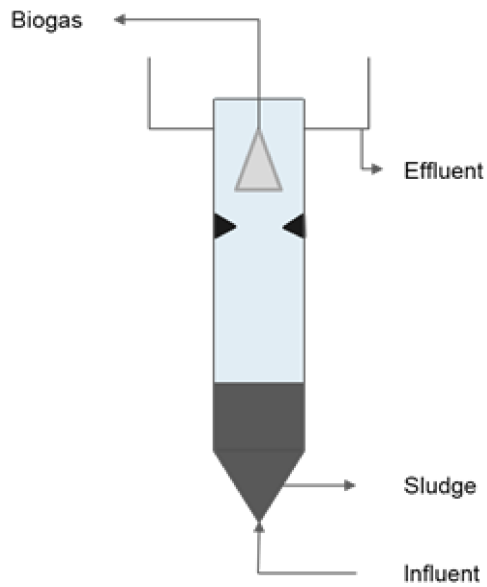

Figure 2.5: Upflow Anaerobic Sludge Blanket (UASB)

Katukiza *et al.* 2014, optimized a filtration system to efficiently treat greywater from an urban slum in Netherlands. Different filtration units in series as well as parallel at constant Hydraulic Loading Rates (HLR) were tested. 90% COD and 94% TSS removal at an HLR of 20 cm/day running the 2 filter columns in series with the GAC filters was reported. The filters were required to backwash every two months while running them in series with pre-treated greywater to enhance filter performance (Katukiza *et al.* 2014). Couto *et al.* 2015, presented a pilot scale study to treat source separated greywater with an anaerobic filter to facilitate reuse applications. The system was able to remove 73% BOD, 72% COD, and 77% TSS from the influent. Post treatment of the effluent with UV disinfection was also done to remove pathogens for prolonged storage duration before reuse (Couto *et al.* 2015).

Microbial Fuel Cell (MFC) has also been used to treat greywater. Sajithkumar and Ramasamy 2015, constructed two low-cost dual chamber MFC's using

PET bottles of 400 mL as anode and cathode chambers and carbon brushes as electrodes with a salt bridge connection between them. At an HRT of 5 Days 77.6% of COD was removed of greywater. The authors claimed a power yield of 0.4 mW/ kg of COD removed (Sajithkumar and Ramasamy 2015). It seemed that their aim was to build an economical MFC, rather than efficient treatment of influent. In a latest development, Khuntia, Chandrashekar, *et al.* 2019, treated greywater in a multi-chambered Anaerobic Biofilm Reactor (AnBR). AnBR was fabricated with 3 chambers. Bottom chamber held sludge, middle chamber held fluidized PVC spirals to support biofilm, and the topmost chamber had multi-use retted coconut fiber for biofilm support, filtration as well as barrier for outflow of sludge, fluidized media and biofilm. The total volume of AnBR was 10 L. AnBR was operated with and without re-circulation of effluent in an up-flow method. A COD removal of 32% was reported without recycling the effluent, whereas a removal 64% COD was achieved with 30% effluent recycling by volume/day (Khuntia, Chandrashekar, *et al.* 2019).

Physical treatment methods for greywater were also recently explored by a few research groups. In the year 2009, Gulyas et al., proposed one of the first post-treatment systems for biologically pre-treated greywater without the use of chemicals. Photocatalytic oxidation and adsorption by GAC in series was reported to have achieved a Total Organic Carbon (TOC) of ≤ 2 mg per L in biologically pre-treated with an initial TOC of ≥ 5 mg/L. It was also concluded that the targeted TOC of ≤ 2 mg/L was not achieved if GAC was added to the same reactor where UV oxidation was performed as photocatalytic oxidation generated polar transformation by-products in the presence of adsorptive material such as GAC (Gulyas et al., 2009). Slow sand, slate filters and GAC treatment of greywater were investigated by Zipf *et al.* 2016 at different filtration rates of 2 and 6 m^3/ m^2/ day. Interestingly, the authors did not encounter much change in treatment efficiency with change in filtration rates even after 28 weeks of operation. Most noticeable achievement of the system was 70% removal of surfactants via the filtration units. However, a marginal removal of COD and BOD of about 56% was achieved. 61% of total coliforms were also reported to have been eliminated by filtration only (Zipf *et al.* 2016). Wang *et al.* 2019, studied the treatment of synthetic greywater by forward osmosis membrane system. A high rejection of nitrate (95.7%), ammonia nitrogen (98.8%), total nitrogen (97.4%) and anionic surfactant (100%) was reported by the researchers. It was also determined that 40°C was the most optimum temperature for the operation of forward osmosis process to treat greywater (Wang *et al.* 2019)

2.3.3 Hybrid treatment

Researchers have used a mixture of aerobic, anaerobic and physical treatments to either have better effluent qualities, achieve cost effective treatment or recover energy or nutrients from the treatment process. Ciabattia *et al.* 2009 investigated a heavily polluted greywater stream from an industrial laundry and used a series of chemical and physical treatment units to bring the effluent to reuse standards in Italy. The prototype system consisted of screening, coagulation, flocculation and dissolved air flotation, sand filtration followed by ozonation and GAC filtration. Part of the GAC filtration effluent was treated with ultra-filtration membrane as well. The effluent quality was immaculate and met stream discharge standards with GAC filtration itself and with an addition of ultra-filtration membrane the effluent met reuse quality standards of Italy w.r.t pH, conductivity, COD, TSS, surfactants and nutrients present in the effluent.

Hernández Leal *et al.* 2010, tested a hybrid system in which they treated greywater by combining anaerobic (UASB) and aerobic (SBR) technologies. Though, an 89% COD removal was achieved by the combined process, aerobic treatment was suggested to be of more advantageous in treating greywater as it removed 97% surfactants as well as 90% COD. On the other hand, authors also suggested a net energy production of 14 kWh/y with the combined treatment system which made it an interesting option to be considered for source separated greywater treatment (Hernández Leal *et al.* 2010). Another hybrid system was introduced by Araneda *et al.* 2018, in which they combined CW with MFC technologies. Removal potential for soluble COD was 91.5%, for TSS was 78.4 % and for nitrate was about 86%, a little lower than CW alone as anode and nitrate competed to act as electron acceptors. For increasing electricity generation potential, the authors provided potentio-static assistance at anode of about -150 mV vs. Ag/AgCl to report a maximum power density of 719.57 ± 67.67 mW m $^{-3}$ (Araneda *et al.* 2018).

Another GAC-electrochemical system was proposed by Garcia *et al.* 2018. Experiments were conducted in a flow-through electrochemical reactor with boron doped diamond anode and a stainless-steel cathode with a packed bed of GAC in between the electrodes named as 3D-ELOX system. To investigate individual effect of adsorption, experiment was run with no applied current (open circuit) and for individual electrochemical impact it was run without GAC media (2D-ELOX). A current density of 15 A/m ^{2} was applied in all the experiments except the open circuit one. 2 L of greywater was circulated in batch mode at 35 L/h for 7 hours. The combined treatment of the system was most efficient with complete removal of colour and 82-89% removal of COD, total organic carbon and turbidity followed by the adsorption experiment with 70-76% removal of the above stated parameters. The 2D-ELOX could only achieve 26-36% removal of COD, total organic carbon and turbidity (Garcia *et al.* 2018).

The CW-MFC synergy was tested again by Bolton and Randall 2019, who combined it with biological sand filter to treat greywater. Only handwashing greywater was treated in this study. The reactor consisted of a GAC anode compacted around a steel mesh current collector in a nylon sleeve and a cathode made up of platinum coated carbon paper. The plant species used was native to Africa as it was easily available in the region of study. The experiment was run in a continuous mode with a combined HRT of 2.2 days. The combined system removed 99% of COD, 63% phosphate, 75% nitrate as well as 4 log concentration of E-Coli from the influent. A maximum current density of 35.8 mA m-3 was also recorded for the system (Bolton and Randall 2019).

An interesting hybrid technology combining a UASB system, a charcoal filtration unit and constructed wetland mechanism was studied by Vidanage *et al.* 2020. The integrated charcoal filter constructed wetland (ICFiWet) system is a vertical up-flow cylindrical reactor in which the influent passes through an anaerobic chamber, the granular media up to the subsurface flow wetland. The system is suggested to be used in small communities for a reuse potential for irrigation purposes. With a removal of 67% BOD the effluent was meeting irrigation standards for Sri Lanka, the place of study. Authors also claimed the advantage of conversion of complex organics into simpler forms which resulted in the increase of Phosphorus content in the effluent which is in the beneficial range of 1.9 ppm for irrigation purposes (Vidanage *et al.* 2020).

2.3.4 Summary

The recent trends in water/wastewater and waste treatment technologies have seen a surge in technologies that can help in recovering energy or nutrients or both (Table 2.6 & 2.4). With increasing research focusing on source separated wastewater it has become important to look into hybrid technologies that explores synergy among aerobic, anaerobic physical and chemical technologies together to get best possible results.

Greywater characteristics vary drastically based on the origin and geography (Gulyas and Raj Gajurel, 2004; Kujawa-Roeleveld and Zeeman 2006). The major issue in its treatment is the presence of toxic surfactants, colloidal COD that biologically degrades very slowly and nutrient recovery (Friedler *et al.* 2005; Hernández Leal *et al.* 2010; Lamine *et al.* 2007; Lesjean and Gnirss 2006). This leads to higher HRT durations for their removal and hence more cost and time is invested in the treatment process. Aerobic treatment and adsorption processes have been useful in treatment of surfactants, removal of odour, colour and turbidity (Merz *et al.* 2007). However, anaerobic and electrochemical processes have found use in recovering energy through the disintegration of pollutants (Elmitwalli and Otterpohl 2007; Hernández Leal *et al.* 2010; Sajithkumar and Ramasamy 2015). Table 2.6 & 2.4 summarizes the treatment technologies discussed in the sections above.

Recent advancements such as CWMFC, 3D ELOX, MBfR and UASB-SBR synergy treatment have ability to intrigue researchers to explore the field of hybrid treatments towards sustainable energy production (Araneda *et al.* 2018; Bolton and Randall 2019; Garcia *et al.* 2018; Hernández Leal *et al.* 2010; Vidanage *et al.* 2020; Zhou *et al.* 2020). Hence, the best from individual treatment technologies should be synergized to tackle the challenges in the treatment of greywater as no individual method has proven efficient enough for sustainable and complete treatment of greywater.

Table 2.3: Greywater Treatment Summary 1

Treatment Technology	COD Removal (%)	BOD5 Removal (%)	TSS Removal (%)	Pathogen Removal (%)	Turbidity Removal (%)	Total Nitrogen Removal (%)	Total Phosphorus Removal (%)	Anionic Surfactant Removal (%)	HRT	SRT (Days)	pH	Working Volume/ Flow Rate	Operating Temperature (oC)	Energy Generation	Reference
UASB	64	-	-	-	-	29.8	15.2	24	16 Hrs	93	-	7L	30	Anaerobic Biodegradability: 76 4%	Elmitwalli and Otterpohl 2007
UASB	52.3	-	-	-	-	21.7	17.4	-	10 Hrs	93	-	7L	30	Anaerobic Biodegradability: 76 ± 4%	Elmitwalli and Otterpohl 2007
UASB	52	-	-	-	-	-	20.6	-	6 Hrs	93	-	7L	30	Anaerobic Biodegradability: 76 ± 4%	Elmitwalli and Otterpohl 2007
SBR	80 (total)	-	-	-	-	-	-	-	0.3 Days	-	-	3.6L	20-30	-	Zeeman *et al.* 2008
UASB	42 (total)	-	-	-	-	-	-	-	0.83 Days	-	7.12	5L	20-30	-	Zeeman *et al.* 2008
SBR	90 (total)	-	-	-	-	15	11	97	12 Hrs	382	-	3.6L 32 ± 3	20-30	-	Hernández Leal *et al.* 2010
Combined (UASB +SBR)	89 (total)	-	-	-	-	2	3	-	7Hrs+ 6Hrs	-	-	5L+3.6L	32 3	71.5 NL/m3 methane	Hernández Leal *et al.* 2010
UASB	51 (total)	-	-	-	-	35	28	24	12 Hrs	15	-	5L	32 ± 3	123 NL/m3 methane	Hernández Leal *et al.* 2010

Table 2.4: Greywater Treatment Summary 2

Treatment Technology	COD Removal (%)	BOD5 Removal (%)	TSS Removal (%)	Pathogen Removal (%)	Turbidity Removal (%)	Total Nitrogen Removal (%)	Total Phosphorus Removal (%)	Anionic Surfactant Removal (%)	HRT	SRT (Days)	pH	Working Volume/ Flow Rate	Operating Temperature (oC)	Energy Generation	Reference
UASB	79 (total)	-	-	-	-	-	10.1	-	8 Hrs	-	-	7L	20	56%	Elmitwalli and Otterpohl 2007
Anaerobic Biofilter with UV Disinfection	71 (total)	73	77	99.99	88	60 (Nitrate)	-	-	-	-	7.6 0.31	2.82 m3/day	20-28	NA	Couto *et al.* 2015
MBR	85	94	-	99	98	63	19	97	18 hrs	-	7.6 ± 0.4	3 L	20	NA	Merz *et al.* 2007
MFC	77 (total)	-	99.5	-	-	-	-	-	-	-	6.5 ± 0.3	300 ml	-	307.69 (mW/m^3)	Sajithkumar and Ramasamy 2015
CW-MFC	91.7	-	78.4	-	-	86.5 (Nitrate)	56.3	-	12 Days	-	7.1 ±0.4	10 L	302	719.57 (mW/m^3)	Araneda *et al.* 2018
CW	27	48.6	82	-	97.2	-	-	-	3 Weeks	-	11.5	12-15L	-	-	Saumya *et al.* 2015
RBC	-	95.9	94.8	-	-	74.3	-	-	-	-	7.40.4	200L	-	NA	Abdel-Kader 2013
RBC	60	53	11.07	-	-	-	-	-	1.5 hrs	-	6.23 0.05	54L	-	-	Pathan *et al.* 2011

Table 2.5: Greywater Treatment Summary 3

Treatment Technology	COD Removal (%)	BOD5 Removal (%)	TSS Removal (%)	Pathogen Removal (%)	Turbidity Removal (%)	Total Nitrogen Removal (%)	Total Phosphorus Removal (%)	Anionic Surfactant Removal (%)	HRT	SRT (Days)	pH	Working Volume/ Flow Rate	Operating Temperature (oC)	Energy Generation	Reference
Aerobic Digestion and Disinfection	68	-	88	99	-	-	-	-	5 Hrs	-	6.13	7.9L + 20L	-	-	Teh *et al.* 2015
IFAS	92.5	85.2	90.2	-	-	89.6	86.6	-	-	-	8.01	9L	30	-	Eslami *et al.* 2018
SM-SBR	91	-	-	99.99	<1 NTU	74	-	-	33	360	-	500	-	-	Kraume *et al.* 2010
SM-SBR	95	-	-	99.99	<1 NTU	91	-	-	18	50	-	600	-	-	Kraume *et al.* 2010
SM-SBR	91	-	-	99.99	<1 NTU	72	-	-	18	50	-	600	-	-	Kraume *et al.* 2010
SBR	88.2	92.7	30	-	-	7.4	Inc	-	0.6 Days	10 Days	7.6	5L	-	-	Lamine *et al.* 2007
SBR	80.4	92.7	30	-	-	95.5	Inc	-	2.5 Days	10 Days	7.6	5L	-	-	Lamine *et al.* 2007
SBR	89	-	-	-	14	-	-	-	1 Day	-	7.4 0.1	50L	-	-	Khuntia, Chandrashekar, *et al.* 2019
MMBR	91	-	-	-	56	-	-	-	1 Day	-	7.4 0.1	50L	-	-	Khuntia, Chandrashekar, *et al.* 2019

Table 2.6: Greywater Treatment Summary 4

Treatment Technology	COD Removal (%)	BOD5 Removal (%)	TSS Removal (%)	Pathogen Removal (%)	Turbidity Removal (%)	Total Nitrogen Removal (%)	Total Phosphorus Removal (%)	Anionic Surfactant Removal (%)	HRT	SRT (Days)	pH	Working Volume/ Flow Rate	Operating Temperature (oC)	Energy Generation	Reference
UASB	79 (total)	-	-	-	-	24	24	-	20 Hrs	-	-	7L	18	48% removed COD to Methane	Elmitwalli and Otterpohl 2007
UASB	79 (total)	-	-	-	-	35.6	21.6	-	12 Hrs	-	-	7L	23	63%	Elmitwalli and Otterpohl 2007
RBC & Sedimentation Basin	75	96	82	98.2	98	87	58	-	2 Hrs + 1 Hrs	-	-	15L +7.5L	-	-	Friedler *et al.* 2005
CW-MFC & BSF	91	-	-	99.99	-	63	75	-	2.2 Days	378	8	5L	25	4.33 (mW/m^3)	Bolton and Randall 2019
MBBR	70	59	87.07	97	66	-	12	30	4 Hr	-	7.2	83.3L	-	-	Chrispim and Nolasco 2017
MBR	85	-	-	-	-	20-80	50	-	2 Hr	4 Days	-	35L	-	-	Lesjean and Gnirss 2006

Chapter 3

Methodology

3.1 Greywater feedstock

Influent greywater stock was prepared as per National Sanitation Foundation (International 2011) standards on a weekly basis. The resulting greywater was aimed to have characteristics in the ranges as shown in the Table 3.1.

Table 3.1: Influent Characteristics

Parameter	Range
TSS	88-160 mg/L
COD	445-485 mg/L
pH	7.4
LAS	47-60 mg MBAS/L
Alkalinity	140-160 mg/L of $CaCO_3$
Conductivity	5.04 µS/cm
e-Coli	None

3.2 Greywater treatment experiment

In this study, a two-step integrated process of MEC followed by an aerobic GAC) bio-filter was investigated for greywater treatment (Figure 3.1). Synthetic greywater was also tested separately using GAC bio-filter and the results were compared with combined treatment and other studies.

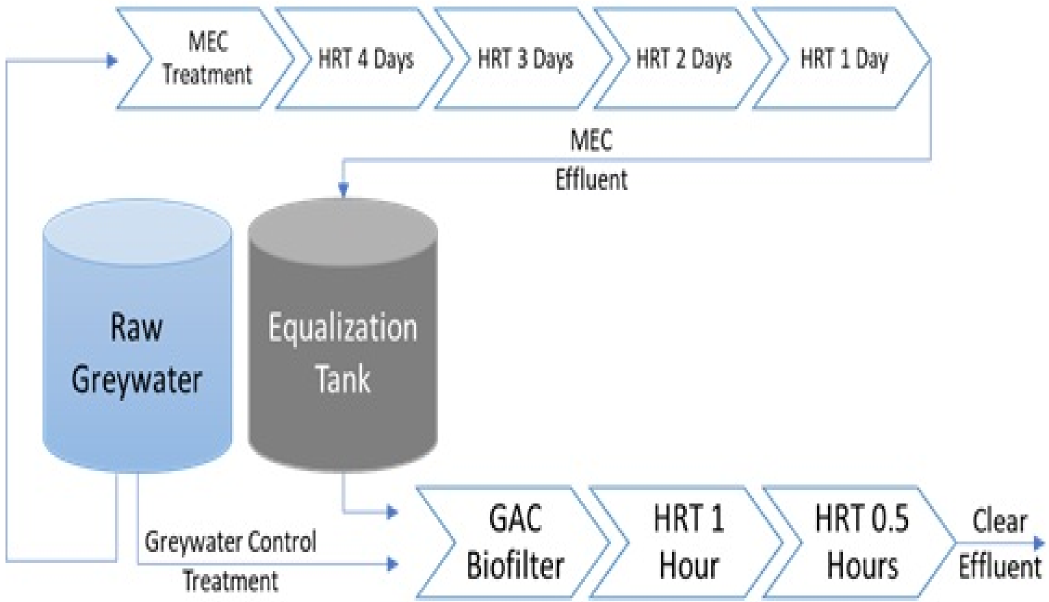

Figure 3.1: Schematic of the Treatment Process

3.2.1 Design and operation of Microbial Electrolysis Cell (MEC)

A dual-chamber MEC was used in this study. MEC was built with plexiglass tubes. The working volumes of anode and cathode chambers were 400 and 200 mL, respectively. Carbon fibres (2293-A, 24A carbon fibre, Fibre Glass Developments Corp., Ohio, USA) attached to a stainless-steel current collector, and a stainless-steel mesh (T304, McMaster-Carr, USA) was used as the anode and cathode electrode, respectively. Carbon fibres were pre-treated as described in the literature (Barua *et al.* 2018). An anion-exchange membrane (AMI-7001, Membranes International Inc., Ringwood, New Jersey, USA) with a projected area of 38·48 cm^2 was sandwiched between the anode and cathode electrodes as a separator. Both anode and cathode chambers consisted of liquid and gas sampling ports. The configuration of the reactor is shown in Figure 3.2.

The anode chamber was equipped with a reference electrode (RE- 5B

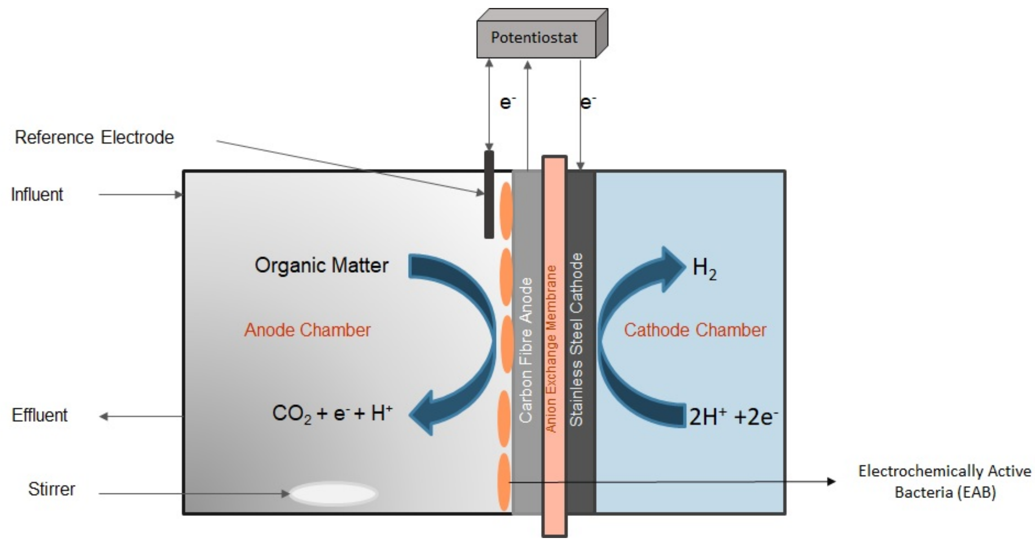

Figure 3.2: Schematic of Microbial Electrolysis Cell

Ag/AgCl reference electrode with flexible connector, Model: MF-2052, Bioanalytical Systems Inc., West Lafayette, USA). It was placed very close (1 cm) to the anode electrode module for controlling the anode potential using a potentio-stat system (Squidstat Prime 4-channel potentio-stat, Admiral Instruments, Arizona, USA). Also, all anode potentials were reported versus Ag/AgCl. Anode potential was set at -0.2 V vs. Standard Hydrogen Electrode (SHE) by the potentio-stat system. The current density was reported based on the volume of the anolyte. The liquid was continuously mixed at 250 rpm with a magnetic stirrer.

For the enrichment of functional anode biofilms, MEC was inoculated with 60 mL of greywater (synthesized as per International 2011) and 60 mL of effluent from an identical mother MEC that had been operated with 25 mM sodium acetate medium for over 24 months. Then, the anode chamber was filled with 280 mL of sodium acetate medium (1600 mg COD/L) supplemented with a nutrient stock solution having specifications as per literature (Barua *et al.* 2018). Before the start-up, nitrogen was purged into the anode chamber for 5 minutes to eliminate any oxygen. The cathode chamber was filled with

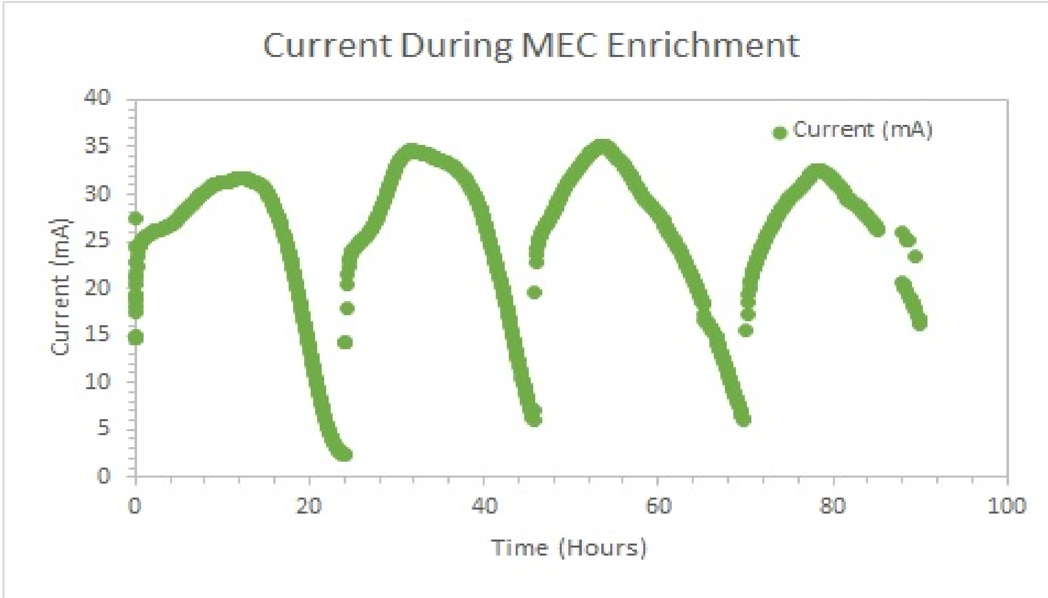

Figure 3.3: Enrichment of MEC with Acetate

tap water (Barua *et al.* 2018). Initially, MEC was operated in batch mode until a positive current density was achieved. Then, it was operated in semi-continuous mode; around 120 mL of anolyte was replaced every day with fresh sodium acetate medium. This process was continued until repeatable peak current densities were achieved (Figure 3.3). Once a stable current density was achieved, 100 mL of anolyte was replaced with fresh greywater every day. This was done for 2 months until repeatable stable peaks of current density were obtained, maintaining an HRT of 4 days. Further, HRT of 3, 2 and 1 days were achieved gradually (total operation time of 45 days) and sampling was done once stable peaks of current were generated for respective HRT's. Samples were collected every 24 hours during the operation with greywater under different operating conditions. These samples were stored at 4°C in a cold room until analysed.

Control experiment for analysing the effect of carbon fiber in the anode electrode was also done wherein, 130 cms of non enriched carbon fiber was placed in 400 ml of greywater for 4 days to assess if adsorption of contaminants occur on the carbon fiber.

3.2.2 Design and operation of aerobic granular GAC biofilter

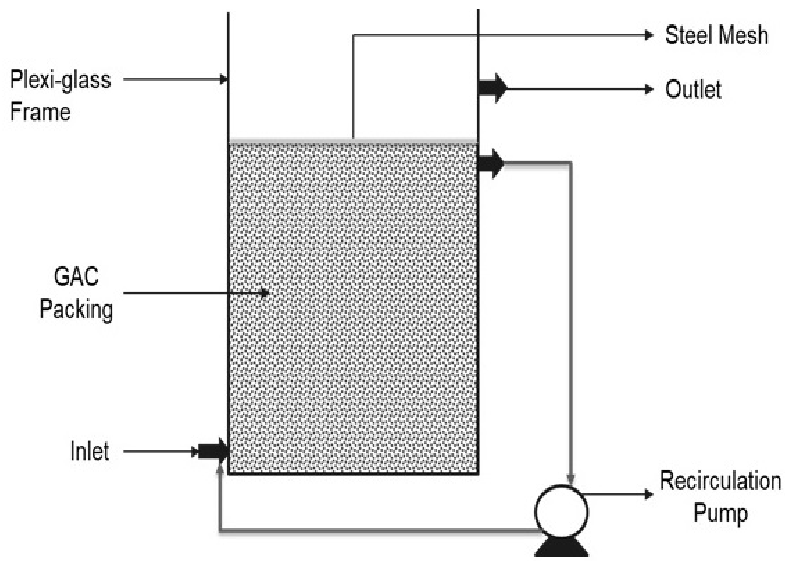

Figure 3.4: GAC-Biofilter Reactor

The MEC effluent was further treated with an up-flow aerobic Granular Activated Carbon (GAC) biofilter (Figure 3.4). The biofilter was built with a cylindrical plexiglass column. The bottom of the column was sealed with a plexiglass plate, and then it was packed with thoroughly washed and oven-dried (105 ° C) GAC. The top of the column was sealed with stainless steel mesh for retention of GAC particles during operation. The working volume of the column was about 400 mL. The liquid inlet port was located at the bottom of the reactor, which was connected to a MEC-effluent storage tank via a feeder pump (Precise peristaltic pump, Model: BT100-2J, Longer precision pump Co., Ltd.). The liquid outlet port of the biofilter was located at the top of the column. There were two additional ports between the liquid inlet and outlet ports that were connected to a pump (Precise peristaltic pump, Model: BT100-2J, Longer precision pump Co., Ltd.) for continuous circulation of liquid within the reactor. The auto-feed pump would feed 165 mL of MEC effluent to the GAC reactor, and the circulation pump would circulate the feed for a total contact period of 30 or 60 minutes. Samples were collected at the outlet at the end of the 30 or 60 minutes contact time and analysed.

Synthetic greywater was also treated through the GAC bio-filter with the contact period of 30 minutes and 60 minutes. The greywater feed remained 165 mL per sampling cycle. To compare the efficiency of biofilm in the biofilter control tests were also done by replacing enriched GAC with fresh GAC without any inoculation in the biofilter. Effluent samples were taken and analysed after the respective contact periods.

3.3 Analytical methods

COD concentration was measured using Hach COD reagent kits (High Range, 20–1500 mg COD/l; Hach Co., Loveland, Colorado, USA). Total Alkalinity was measured using Hach TNT vial tests (Hach, USA). The initial and final pH values of effluent liquid were measured with a benchtop pH meter (Accumet AR15, Fisher Scientific, Pittsburgh, Pennsylvania, USA). Conductivity was measured using an electrical conductivity/temperature meter (Extech EC100, ITM Instruments INC., Edmonton, AB, Canada). The concentrations of different Volatile Fatty Acids (VFA) (e.g., acetate, propionate, butyrate, etc.) were analyzed using an ion chromatograph (Dionex™ ICS-2100, Thermos Scientific, USA) equipped with an electrochemical detector (ECD) and microbore AS19, 2 mm column. For analysis of VFAs, samples were filtered through 0.45 µm membrane syringe filters. Suspended solids concentrations (TSS and VSS) were measured according to standard method (WPCF 2005). Coulombic efficiency (CE) of the reactor was calculated as explained by Zhao *et al.* 2016.

The concentration of LAS in the sample was determined by methylene blue spectrophotometric method. For analysis, 2 mL of liquid sample was diluted to 50 mL with a stock LAS solution (100x) and added into the separating funnel of volume 200 mL. Using phenolphthalein as indicator, drop by drop addition of 1N NaOH solution was done to the separating funnel until the solution colour changed to purple, and then 1N H_2SO_4 was added drop by drop until the purple colour disappeared. Then, 12.5 mL of methylene blue (300 mg/L) was added along with 5 ml of chloroform. The solution was allowed to react with vigorous shaking for 3 minutes by hand and then let sit until the phase separated. The chloroform at the bottom of the separating funnel was transferred into an empty separating funnel of 200 mL volume. The extraction process was repeated twice with 5 mL of chloroform. After that, 25 mL of wash solution was added to this extracted chloroform and vigorously shaken for 30 seconds and let settle until liquid phases separated (Wash solution was prepared by adding 41 ml of 6N H_2SO_4 to 500 mL of water in a 1000mL Flask to which 50g of $NaHPO_4.H_2O$ was added, and the solution was then diluted to 1L). Chloroform separated at the bottom was collected in a volumetric flask through a funnel with a plug of glass wool. The wash solution was again extracted twice with the addition of 5 mL chloroform each time. Then all the extracted chloroform was diluted to 50 mL with additional chloroform. Absorbance was then determined in a spectrophotometer (DR-3900, Hach-USA) at the wavelength of 652 nm against a blank of chloroform. Previously, a five-point calibration curve was made from standard LAS solutions at concentration levels between 0 and 2 mg/L using the steps stated above. From this calibration curve, apparent micrograms of LAS corresponding to the measured absorbance was determined and reported as LAS mg/ L of sample.

Chapter 4

Results and discussion

4.1 Treatment of greywater in MEC

4.1.1 Current density and Coulombic efficiency

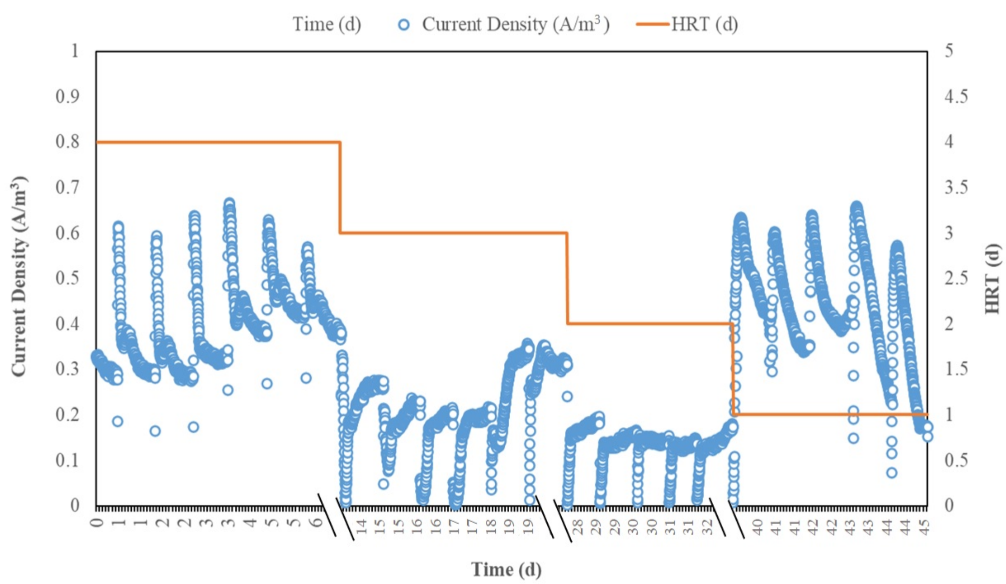

Figure 4.1: Volumetric Current Density in MEC at Different HRTs

Initially, the MEC reactor was operated until a steady state was achieved with GW at an HRT of 4 days. The reactor was operated at a steady state for 2 weeks before the start of the sampling period. The peak current density at an HRT of 4 days was 0.66 A/m^3. However, the current generation dropped to a maximum of 0.35 A/m^3 at an HRT of 3 days and further to 0.2 A/m^3 at an HRT of 2 days. Thus, changes in volumetric current densities deviated from a typical Monod pattern (Dhar, Y. Gao, *et al.* 2013), indicating that fermentation of organics in GW would be required prior to anodic oxidation by Electrochemically Active Bacteria (EAB) and subsequent extracellular electron transfer to the anode. However, at HRT 1 day, the peak current density reached 0.65 A/m^3, which is comparable to the value at HRT 4 days (confirmed by obtaining statistical p value= $9.125*10^{-41}$). Interestingly, COD removal efficiencies did not change after decreasing the HRT from 2 days to 1 day (discussed in Section 4.1.2). The operation of MEC at shorter HRT can lead to the washout of potential competitors of EAB (e.g., acetoclastic methanogens) (Schmidt *et al.* 2013, Asztalos and Kim 2015), which could possibly explain the high current generation at HRT of 1 day. Nonetheless, current density profiles at different current densities demonstrated stable performance of MEC throughout the operating period (Figure 4.1).

Based on an extensive literature search, only three studies could be found on microbial bio-electrical cell systems for greywater treatment, and all of them used MFCs. For instance, the treatability of greywater in microbial electrochemical systems was first investigated by Sajithkumar and Ramasamy 2015. At an HRT of 5 days, their dual-chamber MFC produced a peak current density of 0.15 A/m^3. A recent study by Bolton and Randall 2019 also reported a low current density of 0.035 A/m^3 for an integrated process of constructed wetland MFC and biofilter process. Based on our knowledge, this study first reports the application of MEC's for greywater. The maximum current density observed in this study is 0.65 A/m^3 at an HRT of 1 day. As summarized in Table 4.1, the results suggest that MEC could provide superior current density over MFCs operated under relatively longer HRTs and concentrated greywater

(Sajithkumar and Ramasamy 2015). MEC can provide a favorable metabolic condition for electroactive bacteria due to better process stability achieved through continuous applied voltage/potential. Although the low current density is quite expected for diluted wastewater treatment in MEC (Table 4.1), future research should focus on improving the current density from greywater. For instance, developing multi-electrode MECs could be considered in future studies (Dhar, Ryu, *et al.* 2016). Nonetheless, diluted wastewater fed MECs producing low current density have been successfully demonstrated for on-site generation of value-added chemicals, such as hydrogen peroxide synthesis (Sim *et al.* 2015). Hydrogen peroxide can be utilized for the disinfection of treated greywater before reuse (Chung *et al.* 2020).

Table 4.1: Comparison of greywater treatment in hybrid microbial electrochemical systems

System	HRT (Days)	COD removal (Percentage)	Surfactant removal (Percentage)	Current density or power density (A/m^3 or mW/m^3)	Reference
MEC-GAC-Biofilter	1.04	99.4%	99%	0.66 A/m^3	This study
MFC	5	77.6%	N.A	0.15 A/m^3	Sajithkumar and Ramasamy 2015
CW-MFC	16	91.7%	N.A	N.A	Araneda *et al.* 2018
	2	N.A	N.A	719 mW/m^3 (with potentiostatic assistance) and 33 mW/m^3 (without potentistatic assistance)	
CW-MFC-Biofilter	2.2	99%	N.A	0.035 A/m^3	Bolton and Randall 2019

4.1.2 Organics removal

Figure 4.2 shows effluent TCOD concentrations and corresponding removal efficiencies. The influent COD concentration was maintained 445-485 mg/L throughout the operating period. The Total Chemical Oxygen Demand (TCOD) removal efficiencies at an HRT of 4 days was 58.4%. The COD removal efficiency at 3 days HRT (54.7%) was comparable to 4 days HRT. After reducing the HRT to 2 days, COD removal efficiency decreased to 34.4%, which was further decreased to 31.7% at an HRT of 1 day (Figure 4.2). Thus, COD removal efficiencies were fairly inconsistent with current densities observed at different HRTs. At HRT of 4 days, the highest COD removal efficiency of 61.7% corroborated with the highest peak current density observed among different HRTs. Through the control test as explained in section 3.2.1, we observed no action of adsorption by the carbon fiber in the reactor for the removal of COD. Despite comparable COD removal efficiencies observed for HRTs 1-2 days, peak current density was considerably higher for 1-day HRT. These results suggest that a large percentage of the electrons were lost through pathways other than extracellular electron transfer to the anode by ERB. Various pathways for electron losses may include biomass synthesis, methanogenesis, etc. (Patil *et al.* 2012, Kato *et al.* 2012). The VFA concentration in GW is quite low and was only present in the form of acetate at marginal concentration of 6.5 mg/L. The MEC effluents at different HRTs also showed very minimal accumulation of acetate (<5 mg/L).

4.1.3 Surfactants removal

Surfactants have the ability to break surface tension in a liquid even if present in small quantities. Hence, creating problems of foaming and inhibiting microbial activity. Specially in the biological treatment of wastewater, their toxic nature comes with the affinity for cellular membranes and the capacity to be fixed to certain enzymatic proteins causing inhibition (Aloui *et al.* 2009). Hence, it is accepted that the removal of surfactants from greywater is an important indicator of greywater treatment efficiency. Figure 4.3 shows an-

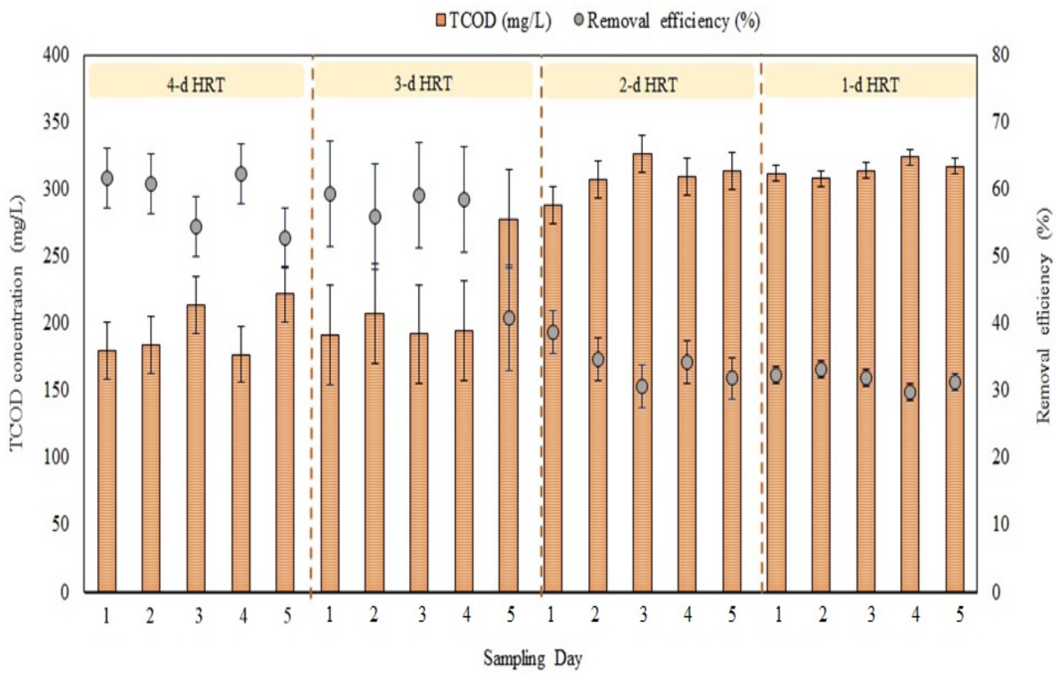

Figure 4.2: TCOD removal by MEC at different HRTs

ionic surfactants (as LAS) removal efficiencies observed at different HRT. The highest LAS removal efficiency of 59.7% was achieved at an HRT of 4 days. After decreasing HRT to 3 days, the removal efficiency remained almost the same (55.6%). However, removal efficiencies decreased with a further decrease in HRT. The average surfactant removal efficiencies were 44.1% and 39.7% at HRT of 2 days and 1 day, respectively.

As AEM was used in the dual-chamber MEC, it was speculated that the anionic surfactants might have been travelling from the anolyte to the catholyte to maintain charge neutrality. Hence, after the completion of all the experiments we analysed the catholyte for the surfactants which might have been accumulated over time. No traces of surfactants were observed in the catholyte. Secondly, the possibility of surfactants being adsorbed on the carbon fiber anode was also investigated with a control test as mentioned in the section 3.2.1. The results of the test indicated that surfactants were not adsorbed on the carbon fiber. Hence, ruling out adsorption on the carbon fiber as the removal mechanism for anionic surfactants in the MEC reactor.

A previous study reported poor anaerobic biodegradability (35±13) of anionic surfactants in GW (Leal *et al.* 2011). However, the authors evaluated the methanogenic biodegradability under mesophilic condition (35°C) for an incubation period of 30 days. In contrast, the results of this study showed MEC operated at ambient temperature could provide superior anaerobic surfactants degradation efficiencies (39.7-55.6%) under HRT of 1-4 days. Surprisingly, surfactant removal efficiency is the most under reported performance parameter. Although a few studies investigated GW traceability in microbial electrochemical systems, none of them reported surfactant removal efficiencies. However, a recent study by Hwang *et al.* 2019, reported that the addition of anionic surfactants could enhance the bio-availability of recalcitrant organics in oily wastewater and enhance electricity generation from MFCs.

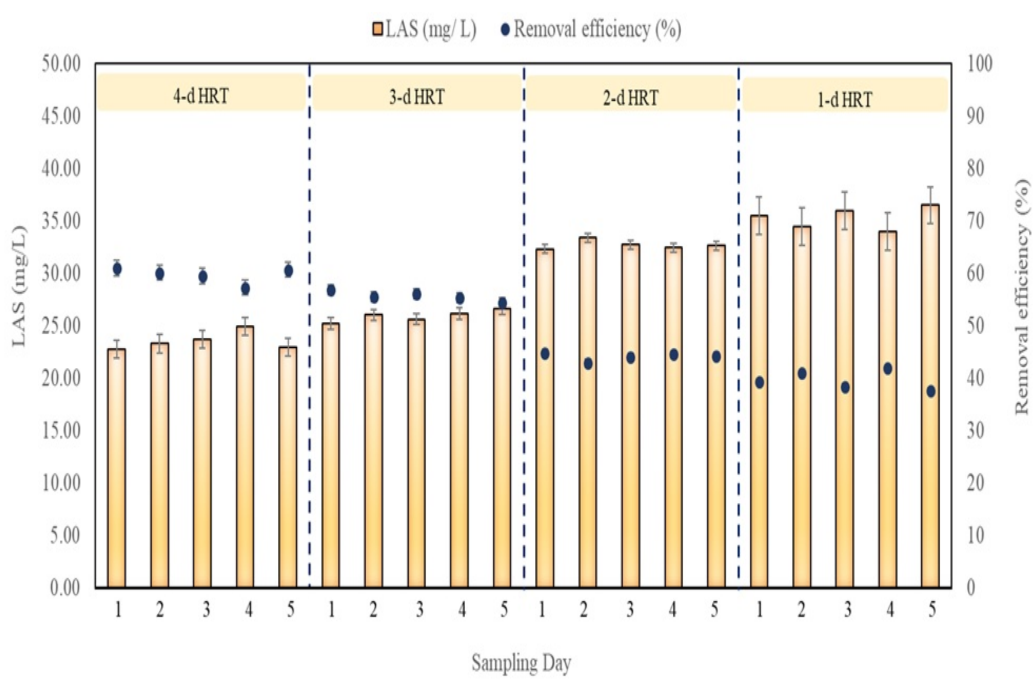

Figure 4.3: Removal and concentration of surfactants by MEC at different HRT

4.2 Performance of GAC biofilter

4.2.1 Treatment of MEC effluent

Although the HRT of 1 day showed superior current density, the effluent COD and SS concentrations were 315 ± 5.36 mg/L and 45 ± 2.2 mg/L, respectively. For the purpose of reuse, Health Canada recommended COD and TSS concentrations to be below 10 mg/L (Drinking Water 2010). Therefore, a polishing step to remove the residuals is necessary to meet optimal effluent quality for reuse. Hence, MEC effluent from 1-day HRT was further treated in a GAC biofilter for very short HRTs (0.5-1 h).

Table 4.2 summarizes the performance of the GAC biofilter. For both HRTs, GAC biofilter provided effective COD removal efficiencies. For instance, the COD removal efficiency was as high as 99.4% at an HRT of 1 h; the effluent COD concentration was only 4 mg/L. The average SS concentration in the final effluent was 9 mg/L. The GAC biofilter reactor was highly efficient in the removal of surfactants, possibly due to its high adsorption capacity (Schouten *et al.* 2007); the effluent anionic surfactant concentration was only 0.74 mg/L. After further decrease HRT to 0.5 h, COD removal efficiency was 98.4% with an effluent COD concentration of 8 mg/L. The average effluent SS concentration for this condition was 12 mg/L. Moreover, the effluent concentration of surfactants increased to 1.88 mg/L. For both HRTs, acetate concentration was below 1 mg/L. Overall, these results suggest that HRT of 1 h would be required for GAC biofilter to adequately polishing MEC effluent to meet recommended guidelines (TCOD and SS) for reuse. Moreover, the effluent did not have any characteristic smell of surfactants and did not form any foam on constant shaking, indicating the efficiency of the combined MEC-GAC biofilter treatment.

Compared to other hybrid bio-electrochemical processes, MEC followed by GAC biofilter in this study demonstrated potential of high-rate treatment system in terms of COD removal; comparable organics removal efficiency was achieved at relatively shorter HRT (see Table 4.2). For instance, the HRT of a constructed wetland MFC-sand biofilter process investigated by Bolton and Randall 2019 was 2.2 days. Their system achieved COD removal efficiency of 99%. Comparable effluent concentration was achieved in this study at an HRT of 25 hours.

4.2.2 Treatment of raw greywater

Although MEC followed by GAC biofilter showed promising results, GAC biofilter operated under 0.5-1 h HRT was very effective treatment efficiency in terms of organics and surfactant removal. However, it was quite expected based on the previous reports that aerobic biofilter would be effective for GW treatment. Therefore, the treatability of raw GW was further assessed with GAC biofilter as a control test condition (see Table 4.3). At 1-h HRT, the TCOD removal efficiency for raw greywater was found out to be 95.5%, which slightly decreased to 92.4% at 0.5-h HRT. The effluent COD concentrations were 21 mg/L and 35 mg/L respectively for 1-h and 0.5-h HRTs. The final SS concentrations were 17 mg/L and 24 mg/L for HRTs of 1-h and 0.5-h, respectively. Thus, these results suggested that GAC biofilter as a stand-alone process could provide effective treatment of GW, while the deployment of MEC could provide an opportunity for energy recovery. Moreover, the effluent from GAC-biofilter operated with raw GW could not meet the recommended guidelines for reuse (COD: 21±1.5 mg/L; SS: 17±3 mg/L). The concentration of surfactants for 1-h HRT was 5.5 mg/L, which further increased to 24±3 mg/L at an HRT of 0.5-h. The effluents from both conditions formed lather on shaking, which indicated evidence of the presence of surfactants. Thus, a further increase in HRT would be required to meet the recommended effluent quality for reuse.

Table 4.2: Performance of GAC-Biofilter System at different HRT

Sample	HRT (h)	Influent COD (mg/L)	Effluent COD (mg/L)	COD Removal (%)	Influent SS (mg/L)	Effluent SS (mg/L)	Influent LAS (mg/L)	Effluent LAS (mg/L)	Surfactant Removal (%)
Raw GW	0.5	465	35±2	92.40	155	24±3	58.09	7.9±0.1	86.57
	1	465	21±1.50	95.5	155	17±3	58.09	5.5±0.4	90.52
MEC-Effluent	0.50	315 ±5.36	7±1.50	98.40	45 ±2.20	12 ±0.80	39.70 ±0.92	1.88 ±0.18	96.78
	1	315 ±5.36	3 ±1.50	99.30	45 ±2.20	9 ±0.81	39.70 ±0.92	0.44 ±0.13	98.73

Interestingly, the GAC biofilter in this study showed effective GW treatment at shorter HRTs as compared to the previous reports on different biofilter studies. In general, biofilters studied for greywater are operated without any liquid recirculation (Araneda *et al.* 2018, Bolton and Randall 2019). Therefore, the GAC biofilter was further operated with raw greywater without effluent recirculation. The COD concentration in effluents from the GAC-biofilter reactor were 97 mg/L and 82 mg/L for 0.5-h and 1-h HRT, respectively. The LAS concentrations were 23.6 mg/L (0.5-h HRT) and 20 mg/L (1-h HRT). Thus, the effluent quality considerably deteriorated after eliminating recirculation. On further testing of biofilter without recirculation, it was observed that the reactor could achieve comparable effluent quality to that achieved with recirculation when HRT was increased to 3-6 hours.

Thus, the results suggest that the recirculation of effluent would be critical to alleviate mass transfer limitations and promote interactions between contaminants and biofilms as well as the adsorption of contaminants by GAC. To confirm this hypothesis, we performed a control test to understand if biofilm plays a key role in the removal of contaminants. We recirculated raw greywater in a similar reactor keeping all the conditions similar but without biofilm enrichment. It was concluded that for TCOD the biofilm plays an important role to achieve the effluent standards stated in table 4.2. However, for surfactants the majority of removal was done by the adsorption mechanism in the GAC biofilter. Removal efficiency of TCOD was recorded around 83.6% for 0.5 hour HRT and 87.5% for 1 hour HRT in the control experiment. On the other hand, for surfactants the removal efficiency was 82% and 88.2% for HRT's 0.5 hours and 1 hour respectively. These results confirm that the removal of surfactants was majorly due to the adsorption mechanism where as for TCOD it was a combination of biological activity as well as adsorption.

Table 4.3: Re-circulation vs no-recirculation in GAC biofilter

Experiment (Raw Greywater Feed)	HRT (H)	Effluent TCOD (mg/L)	Effluent LAS (mg/L)
Recirculation	0.5	35±2	7.9±0.10
	1	21±1.50	5.5±0.40
No-recirculation	0.5	97±3	23.60±1.80
	1	82±1.50	20±0.50
	3	12±3	2.12±0.15
	6	Not Detected	0.35±0.10

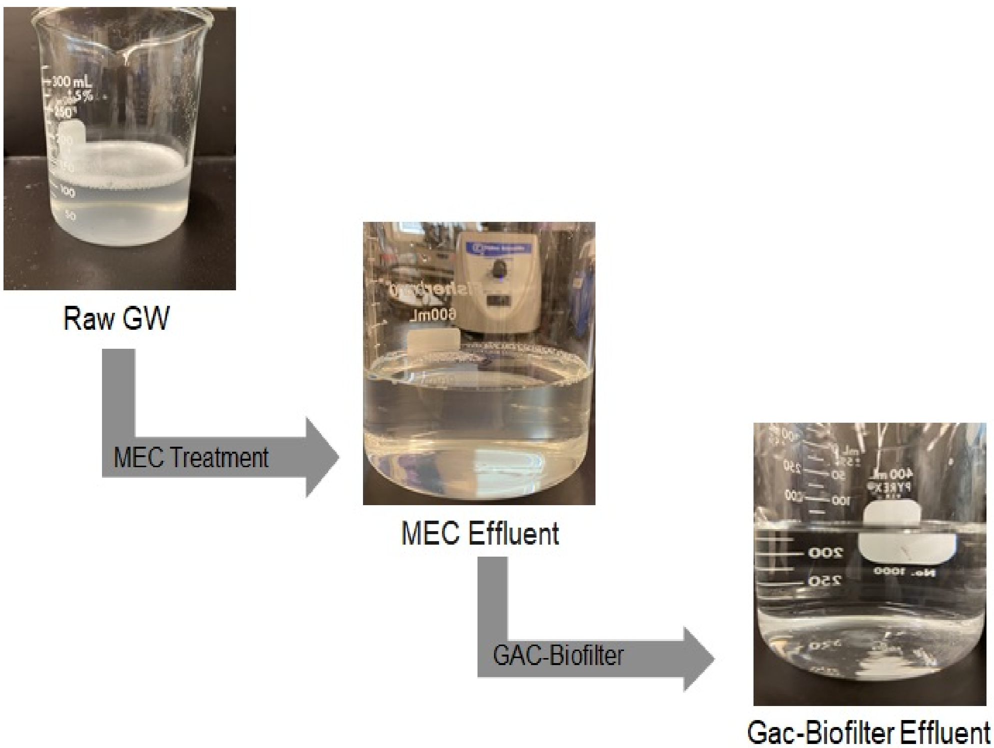

Figure 4.4: Photographs of raw and treated greywater

Chapter 5

Conclusion and recommendations

5.1 Conclusions

A list of major findings in this study is summarized below:

1. The MEC reactor performed depending on the changes in the HRT. The highest organics (as COD) removal of 62% was achieved at an HRT of 4 days. The removal of surfactants was also highest (59.7%) at HRT of 4 days.

2. The MEC reactor was able to generate 0.66 A/m^3 of current density at an HRT of 4 days. The current density dropped at 3-day and 2-day HRT periods. However, analogous to 4-day HRT, the system could generate current density of 0.65 A/m^3 at 1-day HRT possibly due to washout of methanogens.

3. The combined system of MEC-GAC-biofilter could provide about 99% COD as well as surfactants removal with an HRT of only 1.04 days. The recirculation of effluent was found to play a critical role in GAC biofilter operation.

4. The individual performance of the MEC in terms of organics and surfactants was inferior as compared to the performance reported for the

previous aerobic and anaerobic systems used for greywater. However, the system was able to produce higher current density as compared to MFCs previously investigated for greywater. The results indicated that continuous applied voltage/potential during MEC operation was favourable for electroactive bacteria.

5. The combined system could also remove more organics, surfactants and generate higher current density at a much shorter HRT as compared to other studies investigated electrochemical hybrid systems. Although results suggested that aerobic GAC biofilter operated with passive aeration could be a standalone solution for greywater, the potential to recover energy and its subsequent utilization for value-added products could be a great motivation for future investigation and optimization.

5.2 Recommendations

The application of hybrid bioprocess to treat greywater and recover energy is an attractive approach to recover water for potential non-potable reuse. The limited number of studies in this area of research demand a more work to get the best out of these systems. Although MEC followed by GAC biofilter showed promising results in this study, future research should focus on further engineering developments towards increasing current density and decreasing HRT in MEC. For instance, the possibility to enhance current generation and substrate utilization kinetics (i.e., reducing HRT) by providing multiple electrodes for anode biofilms needs to be further investigated. Moreover, the system should be further assessed for other aspects (e.g., pathogen removal) for treated water reuse.

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Appendix A

Table A.1: Results for the Control Tests

Sample	Effluent TCOD (mg/L)	Effluent LAS (mg/L)	TCOD Removal (%)	Surfactant removal (%)
Carbon Fiber (HRT-4D)	465±2	58 ±0.4	0%	0%
GAC Filter (HRT 0.5 hr)	58±2.5	6.8±0.6	87.52%	88.27%
GAC Filter (HRT 1 hr)	76±1.5	10.4±0.4	83.65%	82.06%

In the experiments stated in Table A.1: Influent TCOD- 465±2 mg/L and Influent LAS- 58 ±0.4 mg/L.

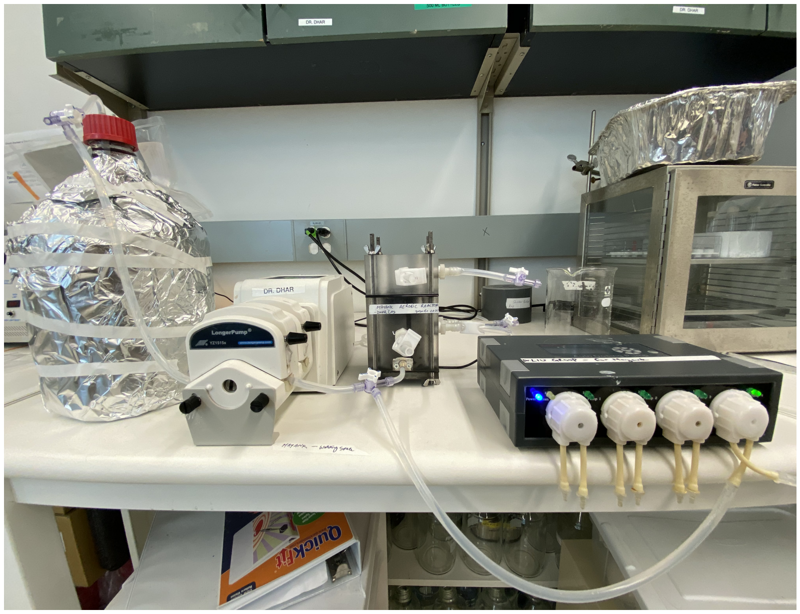

Figure A.1: Image of GAC-Biofilter Setup

Figure A.2: Image of GAC Packaging

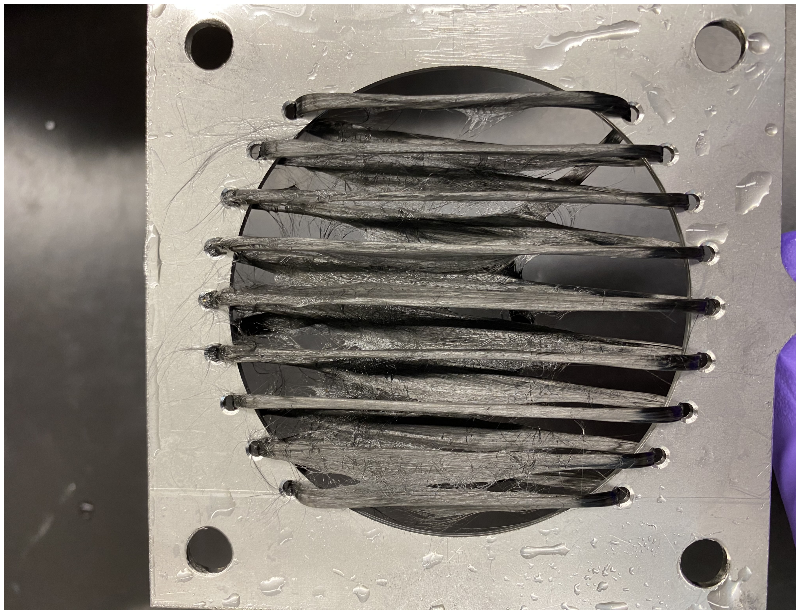

Figure A.3: Carbon Fiber Anode

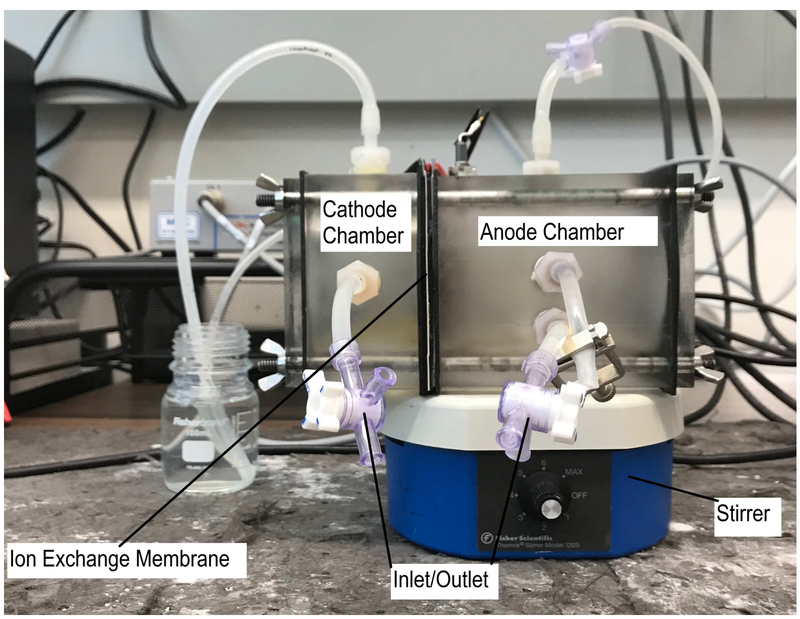

Figure A.4: MEC Reactor

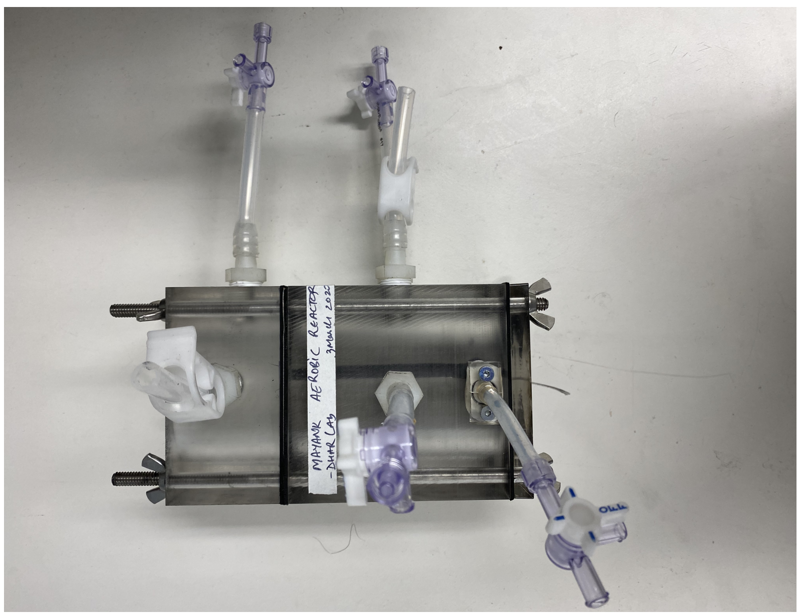

Figure A.5: GAC Biofilter Reactor