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Coupling Microbial Electrolysis Cell and Activated Carbon Biofilter for Source-Separated Greywater Treatment

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Abstract: Reclamation and reuse of wastewater are increasingly viewed as a pragmatic tool for water conservation. Greywater, which includes water from baths, washing machines, dishwashers, and kitchen sinks, is a dilute wastewater stream, making it an attractive stream for extraction of non-potable water. However, most previous studies primarily focused on passively aerated biological and physicochemical treatment processes for greywater treatment. Here, we investigated an integrated process of a microbial electrochemical cell (MEC) followed by granular activated carbon (GAC) biofilter for greywater treatment. The integrated system could achieve 99.3% removal of total chemical oxygen demand (TCOD) and 98.7% removal of the anionic surfactants (linear alkylbenzene sulphonates) from synthetic greywater at a total hydraulic residence time (HRT) of 25 h (1 day for MEC and 1 h for GAC biofilter). For one-day HRT, the maximum peak volumetric current density from MEC was 0.65 A/m^3, which was comparable to that achieved at four-day HRT (0.66 A/m^3). The adsorption by GAC was identified as a key mechanism for the removal of organics and surfactants. In addition, recirculation of liquid within the GAC biofilter was identified as a critical factor in achieving high-rate treatment. Although results indicated that GAC biofilter could be a standalone process for greywater, MEC can provide an opportunity for potential energy recovery from greywater. However, further studies should focus on developing high-rate MECs with higher energy recovery potential for practical operation.

Keywords: greywater; microbial electrolysis cell; granular activated carbon (GAC); biofilter; anionic surfactants

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1. Introduction

Greywater is defined as the used water from all domestic water-related activities (excluding stream generated from the toilet), accounts for 50–70% of the wastewater generated in urban areas [1]. Due to low levels of organics and biological contaminants, greywater is considered as an attractive stream for reclaim, recycle, and reuse for non-potable purposes, including toilet flushing, landscape irrigation, and groundwater aquifer recharge [2–6]. Before being reused, greywater must be adequately treated for its flexible downstream utilization without potential environmental and health risks [7]. To date, various simple to advanced treatment systems, such as biofilter, constructed wetland, and different physicochemical processes (e.g., flocculation, membrane, etc.) have been investigated for greywater [8–13]. Treatment of greywater in biological treatment processes is specifically challenging due to the presence of surfactants [14]. Surfactants, originate from detergents and various personal care products, can break surface tension in a liquid even if present in small quantities [15,16]. The toxicity of surfactants is attributed to their affinity for cellular membranes as well as their capacity to be fixed to certain enzymatic proteins [16]. Thus, the removal of surfactants is a critical concern in greywater treatment.

Among various treatment methods, passive treatment systems like biofilter, constructed wetland, etc., have been reported as a simple and technically efficient method

for greywater treatment [8–13]. For instance, a recent study [17] reported a granular activated carbon (GAC) biofilter that could remove 94% chemical oxygen demand (COD) from greywater for a hydraulic retention time (HRT) of 2.4 h. Interestingly, energy recovery potential from greywater with anaerobic biotechnologies has been rarely investigated. Anaerobic bioprocess, such as anaerobic digestion, is usually known to be more effective for high-strength wastewater [18–20]. However, compared to anaerobic digestion, microbial communities in emerging microbial electrochemical systems, such as microbial fuel cells (MFCs), microbial electrolysis cells (MECs), can provide effective removal of organics from dilute wastewater, including domestic sewage [21–24]. A few studies have suggested that coupling microbial electrochemical systems with passive treatment systems (e.g., biofilter, constructed wetland, etc.) could provide promising results [8,10,25]. For instance, an integrated system of a constructed wetland microbial fuel cell (CW-MFC) and a biofilter could provide 99% COD removal from greywater for an HRT of 2.2 days [10]. Despite previous research efforts in developing these process schemes, an unprecedented advancement is yet to be achieved. Most importantly, high-rate treatment (i.e., reducing HRTs) with bioenergy recovery is desirable for promoting greywater recycling applications. Moreover, the removal efficiencies of surfactants have been unexplored in these previous studies.

Consequently, this study aims at investigating an integrated process of MEC followed by a GAC biofilter for synthetic greywater treatment. Particular attention was given to the removal efficiencies of surfactants in different stages. The system performance was assessed based on the non-potable reuse standards for treated greywater reuse, as suggested by the Canadian federal guidelines [26]. Furthermore, additional experiments were performed to (a) evaluate GAC biofilters performance as a stand-alone process, (b) understand the impact of liquid recirculation on biofilter performance, and (c) understand the organics and surfactants removal mechanisms in both systems.

2. Materials and Methods

2.1. Synthetic Greywater

Influent greywater stock was prepared as per National Sanitation Foundation standards on a weekly basis [27]. The average characteristics of synthetic greywater are as follows: total suspended solids (TSS): 88–160 mg/L, total chemical oxygen demand (TCOD): 445–485 mg/L, linear alkylbenzene sulphonates (LAS): 47–60 mg/L, alkalinity: 140–160 mg/L of $CaCO_3$, conductivity: 50.4 mS/m, and pH: 7.4.

2.2. Bioreactors Setup and Experiments

In this study, a two-step integrated process of MEC followed by a passively aerated GAC biofilter was investigated for greywater treatment. The MEC was operated with greywater under four different hydraulic residence times (HRTs). Based on the current density, 1-d HRT was considered as optimum. Thus, effluent from MEC operated at 1-day HRT was post-treated in a GAC biofilter. As a control condition, the GAC biofilter was further operated with raw greywater, and the results were compared with the integrated treatment scheme (i.e., MEC followed by GAC biofilter).

A dual-chamber MEC was used in this study (see Figure S1, Supplementary Materials). The system was built with plexiglass tubes. The working volumes of anode and cathode chambers were 400 and 200 mL, respectively. Carbon fibers (2293-A, 24A carbon fiber, Fibre Glass Developments Corp., Brookville, OH, USA) attached to a stainless-steel current collector, and a stainless-steel mesh (T304, McMaster-Carr, Chicago, IL, USA) was used as the anode and cathode electrode, respectively. Carbon fibers were pre-treated as described in the literature [28]. An anion-exchange membrane (AMI-7001, Membranes International Inc., Ringwood, NJ, 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 anode chamber was equipped with a reference electrode within ~1 cm of the anode electrode module (RE-5B Ag/AgCl reference electrode with flexible connector, Model: MF-2052, Bioanalytical Systems Inc., (West Lafayette, IN, USA)

for the operation in a three-electrode configuration using a potentiostat system (Squidstat Prime 4-channel potentiostat, Admiral Instruments, Tempe, AZ, USA). During operation, the anode potential was always set at −0.2 V vs. standard hydrogen electrode (SHE). The current density was reported based on the working volume of the anode chamber. The liquid medium was continuously mixed at 250 rpm with a magnetic stirrer.

For the enrichment of functional anode biofilms, MEC was inoculated with 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 a mixture of 60 mL of synthetic greywater and 280 mL of sodium acetate medium (1600 mg COD/L) supplemented with a nutrient stock solution having specifications as per literature [28]. Before the start-up, nitrogen was purged into the anode chamber for 5 min to create anaerobic conditions. The cathode chamber was filled with tap water, where hydrogen gas is produced [28]. Of note, the hydrogen production from the cathode chamber was not monitored during the experiments, as the main focus was on evaluating anodic performance. Moreover, the current generated by the MEC can be converted into various value-added products (e.g., hydrogen, methane, hydrogen peroxide, etc.) by manipulating the cathodic conditions.

Initially, MEC was operated in batch mode until a positive current density was achieved. It was then operated in semi-continuous mode; ~120 mL of anolyte was replaced every day with a fresh sodium acetate medium. This process was continued until repeatable peak current densities were achieved. Once a stable peak current density was achieved with acetate, 100 mL of anolyte was replaced with fresh greywater every day for maintaining an HRT of 4 days. Then, HRTs were gradually decreased from 4 days to 3, 2, and 1 day (total operation time of 45 days). For each HRT condition, sampling was done during the steady-state operating conditions, indicated by similar peak current density for repetitive operating cycles.

The aerobic granular activated carbon (GAC) biofilter was built with a cylindrical plexiglass column (see Figure S2, Supplementary Materials). 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 (Sigma-Aldrich, Saint Louis, MO, USA). The top of the column was sealed with stainless steel mesh for the 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 feed 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 connected to a pump (Precise peristaltic pump, Model: BT100-2J, Longer precision pump Co., Ltd.) for continuous recirculation of liquid within the reactor. During operation, 165 mL of MEC effluent was fed to the GAC biofilter, and then a contact period of 0.5 or 1 h was maintained (depending on the operating conditions). After a contact period of 0.5 or 1 h, samples were collected. As a control condition, the biofilter was also operated with raw greywater for contact periods of 30 min and 60 min. The raw greywater feed volume was the same as the MEC effluent (i.e., 165 mL per operating cycle).

2.3. Analytical Methods and Statistical Analysis

TCOD concentration was measured using Hach COD reagent kits (High Range, 20–1500 mg COD/L; Hach Co., Loveland, CO, USA). Total Alkalinity was measured using Hach TNT vial tests (Hach Co., Loveland, CO, USA). The pH was measured with a benchtop pH meter (Accumet AR15, Fisher Scientific, Pittsburgh, PA, 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 (VFAs) (acetate, propionate, and butyrate) were analyzed using an ion chromatograph (Dionex™ ICS-2100, Thermo Scientific, Waltham, MA, 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 the standard method [29]. The LAS concentrations were determined by the methylene blue spectrophotometric method [30,31]. To evaluate the statistical difference between results from different experimental conditions, we used the student's *t*-test was to test the null (no difference) hypothesis of quality at a 95% confidence level.

3. Results and Discussion

3.1. MEC Performance

3.1.1. Current Density

Figure 1 shows volumetric current densities from MEC under different operating conditions. The peak current density at an HRT of four days was 0.66 A/m^3. However, the peak current densities dropped to ~0.35 A/m^3 at an HRT of three days and further to ~0.2 A/m^3 at an HRT of two days. Thus, changes in volumetric current densities deviated from a typical Monod pattern [32], indicating that fermentation of organics in greywater would be required prior to anodic oxidation by electroactive bacteria and subsequent extracellular electron transfer to the anode. However, at one-day HRT, the average peak current density reached ~0.65 A/m^3, which is comparable to the peak current densities achieved at four-day HRT ($p < 05$). Interestingly, TCOD removal efficiencies did not change after decreasing HRT from 2 days to 1 day ($p < 05$) (discussed later). The operation of *MEC* at shorter HRT can lead to the washout of potential competitors of electroactive bacteria (e.g., acetoclastic methanogens) [33,34], which could possibly explain the high current generation observed at 1-day HRT. Nonetheless, current density profiles at different HRTs demonstrated stable performance of *MEC* throughout the operating period.

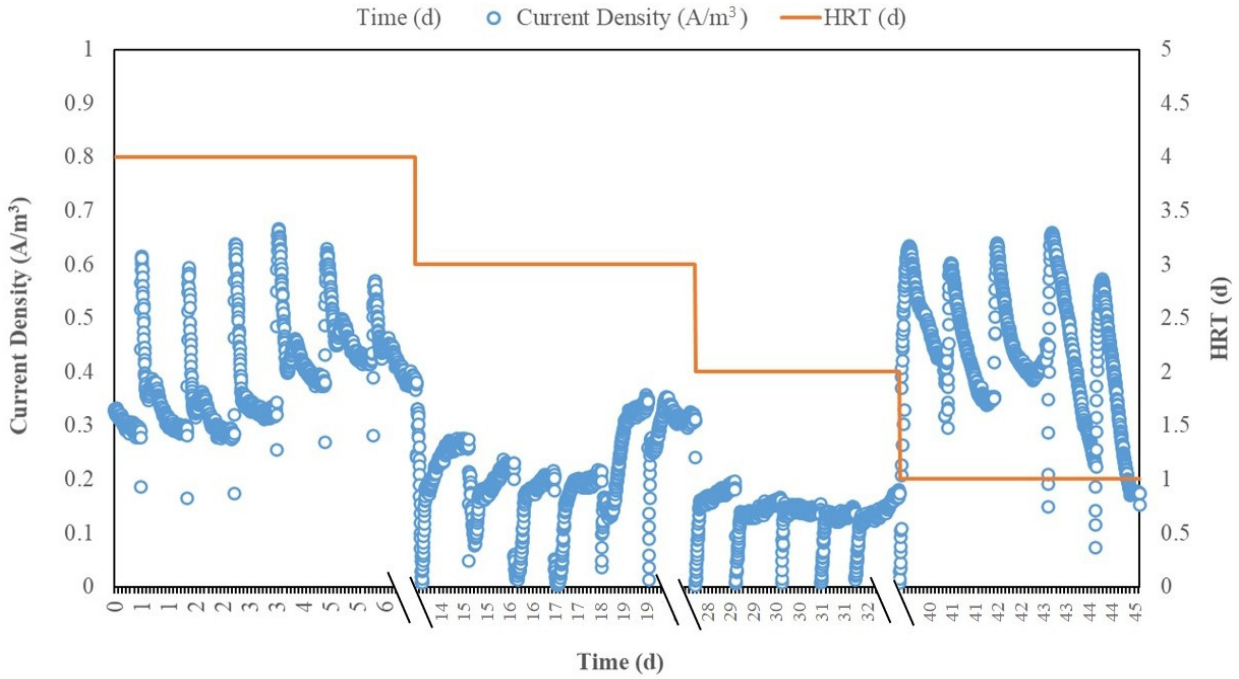

Figure 1. Volumetric current densities from microbial electrochemical cell (MEC) under different hydraulic residence times (HRTs).

Based on an extensive literature search, only three studies could be found on the application of microbial electrochemical systems for greywater treatment, and all of them used microbial fuel cells (MFC) (see Table 1). For instance, the treatability of greywater in microbial electrochemical systems was first investigated by Sajithkumar and Ramasamy [25]. At an HRT of five days, their dual-chamber MFC produced a peak current density of 0.15 A/m^3. A recent study [10], also reported a low current density of 0.035 A/m^3 for an

integrated process of constructed wetland MFC (CW-MFC) and biofilter process. Based on our knowledge, this study first reports the application of MEC for greywater. The maximum current density observed in this study was 0.65 A/m^3 at an HRT of one day. As summarized in Table 1, the results suggest that MEC could provide superior current density over MFCs operated under relatively shorter HRTs. MEC can provide a favorable metabolic condition for electroactive bacteria due to better process stability achieved with continuous applied voltage/potential. Although the low current density is quite expected for dilute wastewater treatment in MEC [24,35], future research should focus on improving the current density from greywater. For instance, developing multi-electrode MECs could be considered in future studies [36]. Nonetheless, dilute wastewater (e.g., domestic sewage) fed MECs producing low current density have been successfully demonstrated for on-site generation of value-added chemicals, such as hydrogen peroxide synthesis [37]. Hydrogen peroxide can be utilized for the disinfection of treated greywater before reuse [38].

Table 1. Comparison of greywater treatment in hybrid microbial electrochemical systems.

Configuration	HRT (d)	COD Removal (%)	Surfactant Removal (%)	Energy Recovery	Reference
MEC-GAC biofilter	1.04	99.4%	99%	0.66 A/m^3	This study
MFC	5	77.6%	–	0.15 A/m^3	[25]
CW-MFC-Biofilter	2.2	99%	–	0.035 A/m^3	[10]
CW-MFC	2	–	–	719 mW/m^3	[8]

3.1.2. Organics Removal

Figure 2 shows effluent TCOD concentrations and corresponding removal efficiencies. The influent TCOD concentration was maintained 445–485 mg/L throughout the operating period. TCOD removal efficiency at an HRT of four days was 58.4%. Almost comparable TCOD removal efficiency of 54.7% was achieved at an HRT of three days ($p < 05$). However, after reducing HRT to two days, TCOD removal efficiency decreased to 34.4%, which was slightly decreased to 31.7% at an HRT of one day. Thus, as discussed earlier, TCOD removal efficiencies were inconsistent with current densities observed at different HRTs. At an HRT of four days, the highest COD removal efficiency of 61.7% corroborated with the highest peak current density observed among different HRTs. Despite comparable COD removal efficiencies observed for HRTs of 1–2 days ($p < 05$), peak current density was considerably higher for one-day HRT (0.65 vs. 0.2 A/m^3). These results suggest that a large percentage of the electrons were lost through pathways other than extracellular electron transfer to the anode by electroactive bacteria. Various pathways for electron losses in MEC may include biomass synthesis, methanogenesis, etc. [39,40]. The VFA concentration in influent greywater was quite low and was only present in the form of acetate (6.5 mg/L). The MEC effluents at different HRTs also showed very minimal accumulation of acetate (<5 mg/L).

3.1.3. Surfactants Removal

Figure 3 shows anionic surfactants (LAS) removal efficiencies observed at different HRTs. The highest LAS removal efficiency of 59.7% was achieved at an HRT of four days. After decreasing HRT to three days, the removal efficiency remained almost the same (55.6%) ($p < 05$). However, LAS removal efficiencies decreased with a further decrease in HRTs. The average LAS removal efficiencies were 44.1% and 39.7% at HRT of two days and one day, respectively. A previous study reported poor anaerobic biodegradability (35 ± 13%) of anionic surfactants in greywater [41]. The authors evaluated the methanogenic biodegradability under mesophilic conditions (e.g., 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 degradation efficiencies (39.7–55.6%) of surfactants under HRT of 1–4 days. It should be noted that surfactants removal efficiency

is the most under reported performance parameter in previous studies. Although a few studies investigated greywater treatment in MFCs (see Table 1), none of them reported surfactants removal efficiencies.

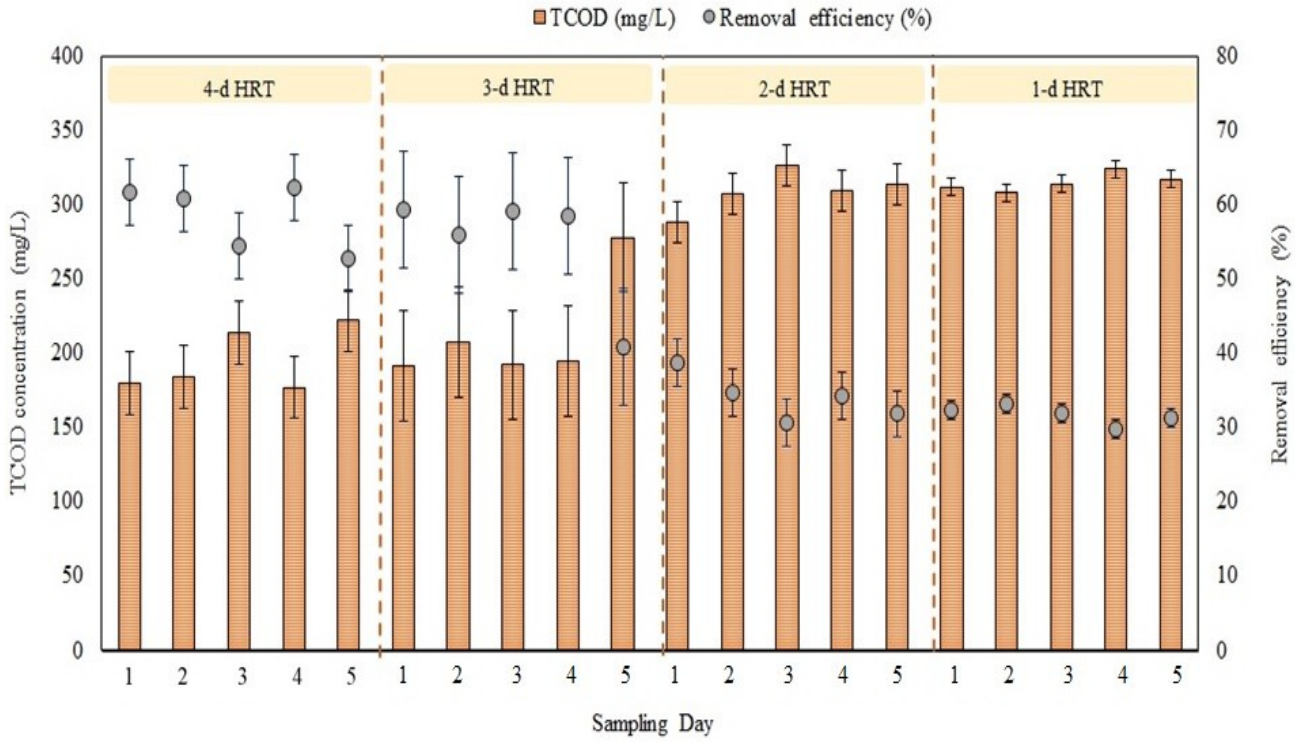

Figure 2. Total chemical oxygen demand (TCOD) concentrations and removal efficiencies from MEC at different HRTs.

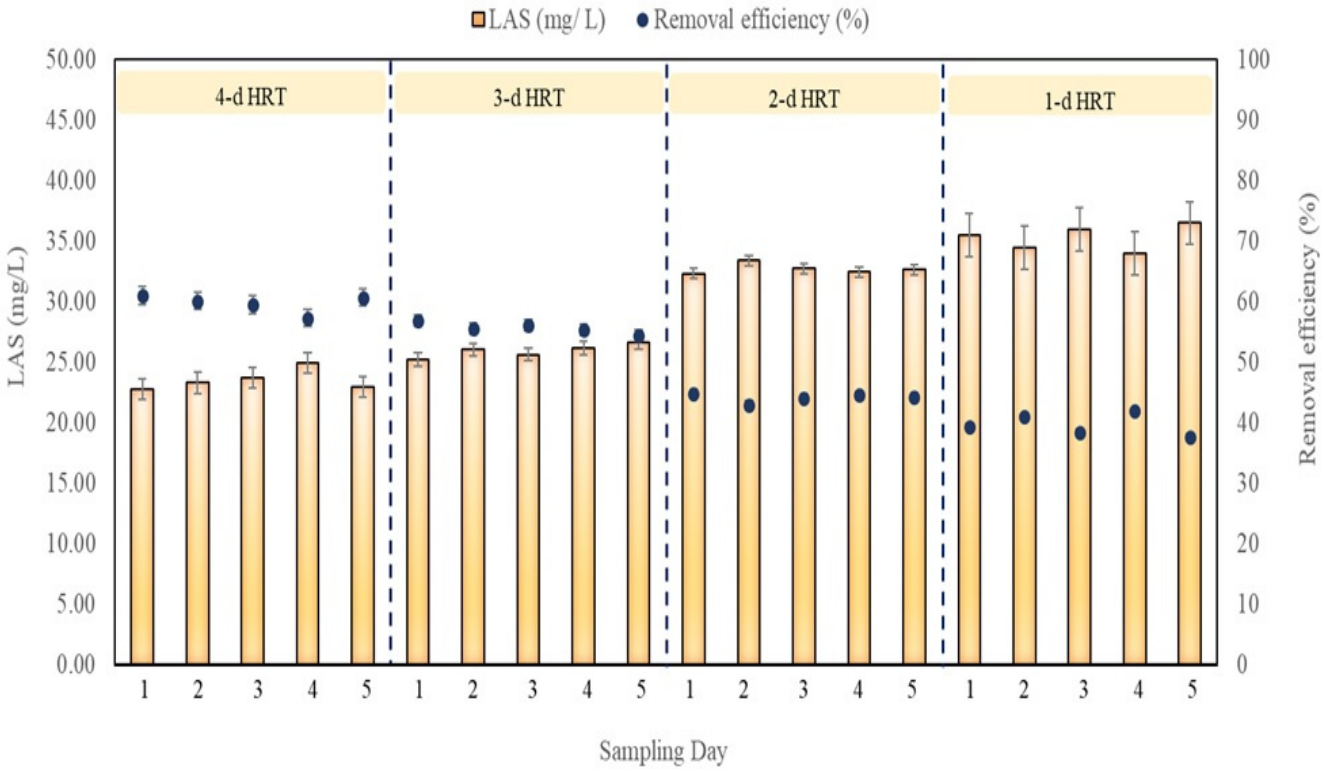

Figure 3. Concentrations and removal efficiencies of anionic surfactants in MEC at different HRTs.

To better understand the LAS removal in MEC in our study, further tests were conducted to evaluate possibilities of LAS adsorption by carbon fibers and transport of anionic surfactants from the anode to the cathode chamber through the anion exchange membrane. To evaluate possibilities of adsorption, a bundle of carbon fibers (130 cm length, without biomass) was placed in 400 mL of freshly prepared greywater (TCOD: 465 ± 2 mg/L, LAS: 58 ± 0.4 mg/L) under anaerobic condition. After four days of incubation, no adsorption of organics and surfactants was observed; TCOD and LAS concentrations remained the same. To evaluate the possibility of surfactant transport from the anode to the cathode chamber, the anode chamber was filled with a mixture of synthetic greywater and acetate medium (volume ratio of 1:1). After four days, liquid samples were collected from the cathode chamber, and no traces of surfactants was observed in the catholyte. Thus, our results suggested that surfactants could be removed by anodic microbial communities, which warrants further investigation as surfactants could also be accumulated in biofilms. Nonetheless, in a recent study [42], the authors reported that the addition of anionic surfactants could enhance the bioavailability of recalcitrant organics in oily wastewater and enhance electricity generation in MFCs.

3.2. Performance of GAC Biofilter

3.2.1. Treatment of MEC Effluent

Although the current densities at one-day and four-day HRTs were comparable, the effluent COD and SS concentrations in the MEC effluent from one-day HRT were considerably higher than that of four-day HRT. Nonetheless, from the energy recovery perspective, one-day HRT would still be attractive. Therefore, the MEC effluent from one-day HRT was further assessed for post-treatment with GAC biofilter. At one-day HRT, 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 TCOD and TSS concentrations to be below 10 mg/L [26]. Therefore, a polishing step will be required to remove the residual organics and suspended solids for meeting the guidelines. Hence, MEC effluent from one-day HRT was further treated in a GAC biofilter for very short HRTs (0.5–1 h). Table 2 summarizes the performance of the GAC biofilter. For both conditions, GAC biofilter provided effective TCOD removal efficiencies. For instance, TCOD removal efficiency was as high as 99.4% at an HRT of 1 h; the effluent TCOD concentration was only 4 mg/L. The average TSS concentration in the final effluent was 9 mg/L. Moreover, the GAC biofilter reactor was highly efficient in removing surfactants, possibly due to its high adsorption capacity [43]; the effluent anionic surfactant concentration was only 0.74 mg/L. After decreasing the contact time to 0.5h, TCOD removal efficiency was 98.4% with an effluent TCOD concentration of 8 mg/L. The average effluent TSS concentration for this condition was 12 mg/L. Moreover, the effluent concentration of surfactants slightly increased to 1.88 mg/L. For both HRTs, acetate concentration was <1 mg/L. Overall, these results suggest that a contact time of 1 h would be required for GAC biofilter to adequately polishing MEC effluent to meet recommended guidelines (TCOD and TSS) for reuse. Futhermore, 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. The photographs of raw and treated greywater from different stages are provided in the Supplementary Information (see Figure S3). Compared to other hybrid microbial electrochemical processes, MEC followed by GAC biofilter in this study, demonstrated the potential of a high-rate treatment system in terms of TCOD removal; comparable organics removal efficiency was achieved at relatively shorter HRT (see Table 1). For instance, the HRT of a constructed wetland MFC-sand biofilter process investigated in a study [10] was 2.2 days. Their system achieved a COD removal efficiency of 99%. Comparable performance (in terms of COD removal efficiency) was achieved in this study at an HRT of 25 h.

Table 2. Performance of granular activated carbon (GAC)-Biofilter System at different HRT.

	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)	LAS Removal (%)
Raw greywater	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

3.2.2. Treatment of Raw Greywater

Although MEC followed by GAC biofilter showed promising results, GAC biofilter as a stand-alone process showed remarkably effective performance in terms of organics and surfactant removal from raw greywater (see Table 2). After a 1-h contact period, the TCOD removal efficiency for raw greywater was 95.5%, which slightly decreased to 92.4% for 0.5-h contact time. The effluent TCOD concentrations were 21 mg/L and 35 mg/L, respectively for 1-h and 0.5-h contact periods. The final SS concentrations were 17 mg/L and 24 mg/L for contact periods of 1-h and 0.5-h, respectively. Thus, these results suggested that GAC biofilter alone could provide effective treatment of greywater. However, the deployment of MEC prior to GAC biofilter could provide an opportunity for energy/resource recovery, which is not possible with GAC as a stand-alone process. However, further research is warranted for the potential practical application of current density produced from MECs treating greywater.

Moreover, the effluent from GAC-biofilter operated with raw greywater could not meet the recommended guidelines for reuse (TCOD: 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. However, the results were quite expected based on the previous reports that aerobic biofilter would be effective for greywater treatment [10,17,44,45].

Interestingly, the GAC biofilter in this study showed effective greywater treatment at shorter HRTs as compared to the previous reports on different biofilter studies for greywater [10,17,44,45]. Based on an extensive literature review, biofilters studied for greywater were operated without any liquid recirculation [8,10,17]. Therefore, the GAC biofilter was further operated with raw greywater without effluent recirculation (see Table 3). The TCOD concentrations in effluents from the GAC-biofilter reactor were 97 mg/L and 82 mg/L for 0.5-h and 1-h contact periods, 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 liquid 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 h. 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 understand relative contributions between biodegradation and adsorption, an identical uninoculated GAC biofilter was operated with raw greywater for contact periods of 0.5 and 1 h with recirculation. The results suggested that a majority of surfactants and organics were removed by adsorption rather than biodegradation (see Table S1, Supporting Information). Thus, the long-term reusability of GAC particles should be investigated in future studies.

Table 3. Impact of liquid recirculation on effluent characteristics from GAC biofilter.

	Contact Time (h)	Effluent TCOD (mg/L)	Effluent LAS (mg/L)
With recirculation	0.5	35 ± 2	7.9 ± 0.10
	1	21 ± 1.50	5.5 ± 0.40
Without 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

4. Conclusions

The results of this study demonstrated that an integrated treatment system of MEC followed by GAC-biofilter could provide ~99% removal efficiencies for both TCOD and anionic surfactants with an overall HRT of 25 h. The recirculation of liquid played a critical role in GAC biofilter operation. Although results suggested that aerobic GAC biofilter can provide a standalone solution for greywater, the potential of energy recovery and its subsequent utilization for value-added products (e.g., hydrogen peroxide) could be a great motivation for future investigation and optimization. Notably, MEC was able to produce higher current density as compared to MFCs previously investigated for greywater. Moreover, the combined system could provide comparable TCOD removal efficiency at a relatively shorter HRT than other studies investigating hybrid microbial electrochemical systems. Nonetheless, further research would be needed to optimize process parameters, scale-up, techno-economic, and life-cycle assessment of the integrated process.

Supplementary Materials: The following are available online at https://www.mdpi.com/2227-9717/9/2/281/s1, Figure S1: Schematic diagram of microbial electrolysis cell, Figure S2: (**a**) Schematic diagram of granular activated carbon (GAC) biofilter, (**b**) photograph of GAC biofilter setup, Figure S3: Photographs of raw and treated greywater, Table S1: Results from uninoculated GAC biofilter.

Author Contributions: M.D., was responsible for experimental design, laboratory experiments, data collection and analysis as well as the manuscript composition. B.R.D. and Y.L. planned and supervised the study. All authors contributed to the manuscript preparation. All authors read and agrees to published version of the manuscript.

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