Impact of Applying Electrocoagulation Pre-treatment Step on Grey Water Treatment by Submerged Membrane Bioreactor

Khalid Bani-Melhem* **and Edward Smith**

Environmental Engineering Program, Department of Construction and Architectural Engineering, The American University in Cairo, Cairo 11511, Egypt.
*(E-mail: kmelhem@aucegypt.edu)

ABSTRACT

Chemical coagulation is widely used for enhancing the performance of various water and wastewater treatment systems, including membrane filtration processes such as submerged membrane bioreactor (SMBR) units. However, the addition of chemicals to the wastewater may result in production of undesirable chemical by-products or increase the volume of sludge in the bioreactor. Alternatively, applying electrocoagulation (EC) technique has been shown to be a promising and novel approach in SMBR applications. In this study, the impact of applying EC step was investigated as a pretreatment stage for grey water treatment by SMBR system. The main objective of the present study was to investigate the performance of the integrated process of EC-SMBR under different voltage gradients. The performance of the integrated process of (EC-SMBR) was compared with the operation of a SMBR system operated alone. The comparison was achieved in two sequential operational phases in which two different voltage gradients were applied in the EC step. In both phases, aluminium electrodes were used. The results indicated that the EC unit can contribute in improving the SMBR performance if an adequate voltage gradient was applied in EC step. The improvement was observed clearly in Phase II when a voltage gradient of 1.26 V/cm was applied. Applying a low level of voltage gradient of 0.64 V/cm (Phase I) in EC step did not exhibit a significant improvement in pollutant removals.

KEYWORDS: Grey water, submerged membrane bioreactor, membrane fouling, electrocoagulation,

INTRODUCTION

In the last several decades, increasing attention has been given for grey water (GW) as a valuable resource for wastewater treatment and reuse. Grey water is any source of wastewater generated from the kitchen, laundry and bathroom (sink, bath and shower) and any other non-toilet household wastewater (Schafer *et al.*, 2006). Grey water constitutes 50-80 percent of the total wastewater generated in households (Friedler and Hadari, 2006) which makes it a good source for water reuse.

In order to be ready for reuse, the GW should be treated effectively. Although physical (March *et al*., 2004) and chemical (Lin et al., 2005, Pidou *et al*., 2008) processes can effectively remove suspended solids, organic materials and surfactants, they are not cost-effective for removing the full array of dissolved components in wastewater (Li *et al*. 2009). The biological processes in combination with physical or physical-chemical processes have been found to be the most efficient methods for grey water treatment (Li *et al*. 2009).

In recent years, the submerged membrane bioreactor (SMBR) has been investigated as an attractive method for grey water recycling since it combines physical separation of colloidal substances, including pathogenic bacteria, together with aerobic biological treatment of dissolved organic matter (Liu *et al*., 2005, Smith and Bani-Melhem, 2012). However, one drawback that limits more widespread application of SMBR units is the decreasing of filtration efficiency with time resulting from fouling of the membrane (Le-Clech *et al*., 2006). Therefore, most of the recent studies on SMBR applications have focused on improving the performance of SMBR to reduce the problem of membrane fouling. In this domain, improving the characteristics of the activated sludge in the bioreactor was considered a potential approach for reducing the fouling in SMBR applications. The strategy of this approach is based on increasing the flocs size of the activated sludge particles by enhanced flocculation in the bioreactor, thereby reducing the contribution of small particles to membrane fouling *via* plugging the membrane pores.

Increasing the floc size of the activated sludge particles has been traditionally achieved by adding chemical coagulants as alum and iron salts (Lee *et al*., 2001; Wu and Huang, 2008). However, the addition of chemicals to the wastewater may result in production of undesirable chemical by-products or increase the volume of sludge in the reactor (Clark and Stephenson, 1998). Alternatively, applying direct current (DC) field on the activated sludge has been shown as a promising and novel approach in SMBR applications (Chen *et al*., 2007; Bani-Melhem and Elektorowicz, 2010; Bani-Melhem and Elektorowicz, 2011; Liu *et al*., 2012a, Liu *et al*., 2012b). This approach can be implemented by integrating electrocoagulation (EC) technology in the same reactor with the SMBR (Bani-Melhem and Elektorowicz, 2010; Bani-Melhem and Elektorowicz, 2011) or using the membrane as a cathode (Liu *et al*., 2012b).

However, applying a DC field directly in the activated sludge reactor may be harmful to microorganism activity (Bani-Melhem and Elektorowicz, 2011). An alternative strategy is to use the EC unit as a pre-treatment step before the SMBR system in order to prevent direct contact of the microbial community with the applied DC field. This latter alternative was utilized in a previous study (Bani-Melhem and Smith 2012); namely, an electrocoagulation unit served as pre-treatment to an SMBR to treat grey water with the two reactor systems in series in continuous flow mode (*i.e*. EC-SMBR process). The results demonstrated that the EC-SMBR may be an effective method not only for improving the overall performance of the membrane filtration process, but also for increasing the quality of the treated grey water over a conventional SMBR. However, ammonia nitrogen (NH_3-N) reduction through biological nitrification was found to be higher in the SMBR than the EC-SMBR, likely due to the sensitivity of the nitrifying bacteria to the excessive aluminium concentration resulting from the high voltage gradient applied in the EC unit.

Accordingly, this study investigates the use of lower values of the applied DC in the EC step so as not to impede biological growth and treatment. Therefore, the main objective of the present study was to explore the impact of applying lower voltage gradients in EC unit as a pre-treatment step on grey water treatment by submerged membrane bioreactor (SMBR).

METHODOLOGY

Experimental Set-up

A laboratory scale experimental set-up was used in this study (Fig. 1). The set-up consisted of a SMBR proceeded by an electrocoagulation (EC) unit. For a comparison purposes, another conventional SMBR process was operated in parallel with the EC-SMBR as a reference system. Each bioreactor had a 3.72 L working volume. Two identical hollow fiber membrane module, ZeeWeed-1, were used in this study. The effluent from each membrane module was withdrawn *via* a peristaltic pumps (E SERIES, Barnant Company, USA) operated at constant transmembrane pressure. The EC unit had a 2.5 L working volume. The anode was a tubular shape with 810.5 cm^2 effective surface area made from a rectangular sheet and mounted to be close to the internal wall of the EC unit while the cathode was a rectangular rod (33.5 cm ×2 cm × 1cm) fixed in the centre of the EC unit. The radial distance between electrodes was set at 6.5 cm. The electrodes were connected to an external DC power supply connected with a timer to supply the DC intermittently (15 min ON / 60 min OFF). A level sensor was connected with the feeding pumps of each process *via* a level controller system to maintain constant volume in each bioreactor. Compressed air, at a flow rate 4.2 L/min, was supplied through air diffusers at the bottom of each membrane module. The compressed air was used for: (1) maintaining the required dissolved oxygen for the microorganisms; (2) creating a shear stress for effective scouring of the membrane surfaces; and, (3) providing good mixing of the sludge suspension in the bioreactors. The grey water in EC unit was rapidly mixed with a stream of air at a flow rate of 2 L/min to enhance mass transfer of aluminium ions away from the anode and achieve a good mixing.

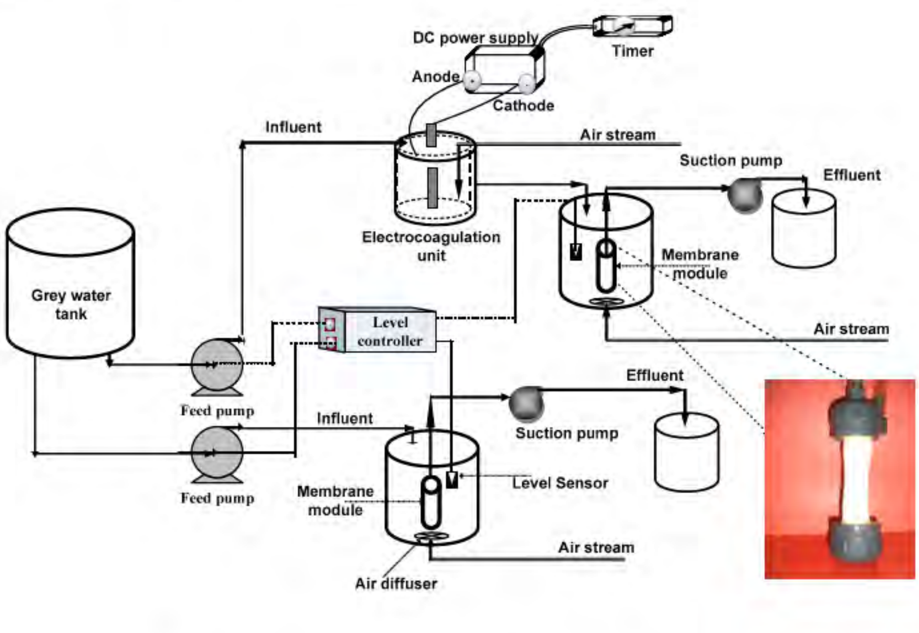

Figure 1. Schematic diagram of the experimental set-up

Grey Water Characteristics

Actual grey water was used in this study. The grey water was collected from one of the facilities operations buildings on the campus of the American University in Cairo (AUC), Cairo, Egypt. Table 1 shows a summary of statistical analysis of the grey water characteristics used in this study during the experimental phases. Both bioreactors were seeded with waste activated sludge (WAS) from El-Jabal El-Asfar municipal wastewater treatment plant in the Cairo prior to the experimental start-up.

Experimental Procedure and Operating Conditions

Table 2 summarizes the different operation conditions under which the two processes were run and monitored. Both processes (EC-SMBR and SMBR) were operated in parallel at room temperature (21.7 ± 0.6 ^{o}C) without control for 60 days comprising two sequential phases (30 days per phase). Each phase was divided into six consecutive filtration-cleaning cycles of 5 days each. The EC unit was operated at 0.64 V/cm and 1.26 V/cm in Phases I and II, respectively.The fouling behaviour was evaluated phenomenalogically by measuring the decline of permeate flux with time. No backwashing of the membrane module was performed during the operation period. After five days of operation and before starting each new cycle, and in order to restore most of the membrane's permeability, the membrane modules were removed from the bioreactors and physical and chemical cleaning were applied.

In order to observe and compare the impact of membrane fouling for the different process conditions, both processes were operated at a constant transmembrane pressure that was created by withdrawing the effluents via a peristaltic pump operated at a constant suction pressure (26 ± 0.5 kPa). Moreover, both processes were operated at complete sludge retention time (SRT); *i.e.*, during the entire experimental period, no sludge was withdrawn from both bioreactors except for the required measurements.

Table 1. Characteristics of grey water used as feed to SMBR and EC-SMBR processes

Water quality indexes	Average value ± Standard deviation (samples' number)	
	Phase I	Phase II
pH	6.60 ± 0.42 (14)	6.19 ±. 0.36 (11)
DO (mg/L)	0.73 ± 1.28 (14)	0.84 ± 0.47 (11)
TDS (mg/L)	254 ± 20 (14)	262 ± 46 (11)
Conductivity (ms/cm)	0.507 ± 0.41(14)	0.524 ± 0.093 (11)
TSS (mg/L)	40 ± 19 (12)	42 ± 12 (12)
COD (mg/L)	650 ± 259 (13)	1038 ± 196 (11)
NH_3-N (mg/L)	3.19 ± 0.91 (13)	4.33 ± 1.27 (11)
TP (mg/L)	1.50 ± 0.47 (13)	1.86 ± 0.46 (13)
Turbidity (FTU)	93 ± 47 (14)	132 ± 26 (10)
Color (PtCo)	505 ± 266 (14)	709 ± 136 (11)
Anionic surfactants (mg/L)	91.05 ± 33.27 (6)	160.8 ± 11.9 (6)
Fecal coliform (CFU/ 100 mL)	26×10^4	14×10^4
Total coliform (CFU /100 mL)	43×10^4	32×10^4

Table 2. Operation conditions of the SMBR and EC-SMBR processes

Item	SMBR	EC-SMBR	
	Phase I and Phase II	Phase I	Phase II
Operation time (days)	60	30	30
Suction pressure (kPa)	26	26	26
Air flow rates to bioreactor (L/min)	4.2	4.2	4.2
SRT (days)	complete	complete	complete
Applied voltages gradient (V/cm)	0	0.64	1.26
DC exposure time ON (min) / OFF (min	-----	15/60	15/60
Average MLSS (mg/L)	3920	3264	3412

Analytical Methods
Influent and effluents were sampled regularly and analyzed by Hach methods (Hach, DR 2000, USA) for COD, ammonia nitrogen (NH_3–N), total phosphate (TP), anionic surfactants (AS), total dissolved solids (TDS), conductivity, color and turbidity. The average values presented in this study were calculated as an arithmetic mean of the collected data. Mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) were performed according to Standard Methods (APHA, 1998). Total and faecal coliforms were determined by the membrane filtration procedure. The dissolved oxygen (DO) concentration was measured using a DO meter (SensIon 8, Hach, USA). The values of pH and temperature were measured using a pH meter model CG 842 (SCHOTT, Germany). The size distribution of activated sludge particles were determined by Horiba Laser scattering particle size distribution analyzer Model LA-300 (Horiba, USA).

RESULTS AND DISCUSSION

Removal of Colour and Turbidity
Figure 2 shows a comparison between the performances of EC unit, EC-SMBR and SMBR processes with respect to the changes in the colour and the turbidity during the two experimental phases. Water permeate quality from the two processes exhibited excellent colour removal in the two experimental phases with an average removal in Phase I close to 91%. The EC step had no significant impact on the colour removal when EC was operated at 0.64 V/cm. However, the effluent colour quality from EC-SMBR was better in Phase II when the applied voltage was doubled with an average value of 98% compared to 93% from SMBR-only process. With respect to the turbidity, the EC step achieved notably better removal in Phase II (44%) compared to the percentage removal in Phase I (29%). This improvement was reflected in the overall removal by EC-SMBR process where the average percentage removal of the turbidity increased from 92% in Phase I to 98% in Phase II versus the turbidity removal by SMBR alone which remained at 92%.

Removal of microorganisms and total suspended solids (TSS)
The UF module used in this study achieved a complete removal of total and fecal coliforms in both processes. Influent total dissolved solids (TDS) and total suspended solids (TSS) concentrations in grey water were relatively steady over the study period. The total suspended solids (TSS) were not detected in effluents of both processes.

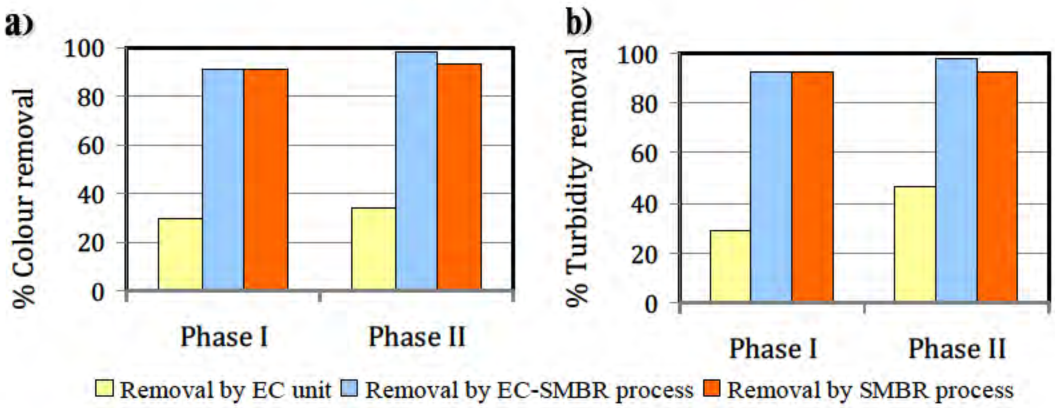

Figure 2. Average percentage removal efficiencies of the EC unit, EC-SMBR process and SMBR alone during the two experimental phases: a) % Colour removal and b) % Turbidity removal

Removal of COD, NH3-N, Total Phosphorus (TP) and Anionic Surfactants (AS)

Figure 3 shows the average percentage removals of COD, TP, NH_3-N and AS achieved by EC unit, EC-SMBR process and SMBR process alone during the two experimental phases. In Phase I, both processes showed the same percentage total removal with respect to COD with an average value closed to 80% (Fig. 3-a), with the EC unit achieving 18% removal of COD. Once again, the output results from Phase I demonstrated that the applied DC had no effect on the SMBR performance removal with respect to COD. In Phase II, the %COD removal increased to 23.5% by EC, increasing the overall performance of the EC-SMBR to 88% while the %COD removal by SMBR alone remained at 80%.

The benefit of integrating EC with SMBR in treatment of the grey water was most clearly observed in the concentration reduction of the total phosphate (TP) in both experimental phases. The EC-SMBR process achieved average removal efficiencies of 73% and 90% in comparison with average removal efficiencies of 54% and 65% achieved by SMBR only throughout the duration of the experiment phases I and II, respectively (Fig. 3-b). This improvement likely resulted from the chemical reactions that occurred in the EC-SMBR process between aluminium ions and soluble phosphorus. When the metal ions (Al^{3+}) began to appear on the anode side ($Al \rightarrow Al^{3+} + 3e^-$) in the grey water solution, it reacted with hydroxide ion (OH^-) produced in the cathodic zone ($3\ H_2O + 3e^- \rightarrow 1.5\ H_{2\,(g)} + 3OH^-$), producing aluminium hydroxide ($Al^{3+}{}_{(aq)} + 3\ H_2O \rightarrow Al\ (OH)_{3(s)} + 3H^+{}_{(aq)}$) in the solution (Kobya *et al.*, 2006). The produced aluminium hydroxide has the characteristic of sweep flocs with a large surface area (Kobya *et al.*, 2006) that is beneficial for the rapid adsorption of soluble phosphorus in the bioreactor.

It is noteworthy that the removal efficiency of TP by EC-SMBR process is better than in other reported studies using MBR systems on grey water treatment without integrating an EC unit. The performance of EC-SMBR and SMBR processes with respect to TP removal was compared with other MBR processes for grey water treatment as shown in Table 3.

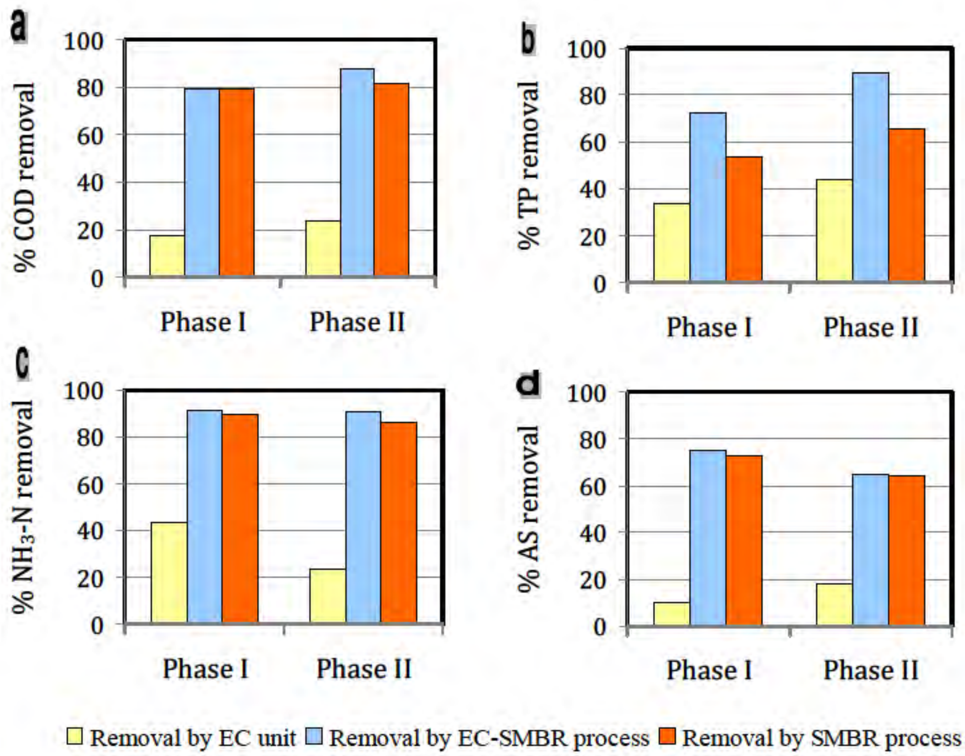

Figure 3. Average percentage removal efficiencies of the EC unit, EC-SMBR process and SMBR alone during the two experimental phases: a) % COD removal, b) % $Po4^{-3}$ removal, c) % NH_3-N removal and d) Anionic surfactants (AS) removal

Table 3. Comparison of TP removal from grey water by SMBR system with other MBR processes

Reference	Average influent (mg/L)	Average effluent (mg/L)	% Removal
Lesjean and Gnirss (2006)	7.4	3.5	53 %
Merz et al. (2007)	1.6	1.3	19 %
Smith and Bani-Melhem (2012)	0.52	0.23	57 %
This study:			
Phase I (without EC)	1.50	0.69	54 %
Phase I (with EC)	1.50	0.53	73 %
Phase II (without EC)	1.86	0.50	65 %
Phase II (with EC)	1.86	0.19	90 %

Ammonia nitrogen (NH_3-N) reduction was excellent in Phase I in both processes (EC-SMBR and SMBR) as the average percentage removals were nearly 92% and 90% for EC-SMBR and SMBR processes, respectively (Fig. 3-c). In Phase II, the average percentage removal by EC-SMBR process was 91.1% while a small decrease was observed in SMBR process (87% on average). On the other hand, it was observed that the % NH_3-N by EC unit alone was actually better in Phase I (average removal = 44% versus 24%) than in Phase II. This may be an indication that the microbial community is sensitive to aluminum concentration (Dorea *et al.*, 2008) which is related to the magnitude of the DC applied in the electrocoagulation step. By placing the processes in series, however, rather than in the same reactor as in previous studies, the overall process performance for NH_3-N conversion is maintained at a high level.

The removal of AS was better in EC step in Phase II (18 %) than Phase I (10 %) which was attributed to the increase in the applied voltage in Phase II (Fig. 3-d). However, the overall performance of both processes with respect to AS removal was better in Phase I (75 % by EC-SMBR and 73 % by SMBR only) than Phase II (65 % by EC-SMBR and 64.4 % by SMBR). This could be attributed to the significant increase in the influent AS concentration during the operation of Phase II (Table 1).

Membrane Permeability

Because the SMBR and EC-SMBR processes in this study were operated on the basis of constant TMP pressure, a decline in permeation flux during the experimental phases is assumed to be due to the fouling phenomenon. At the beginning of each cycle, the membrane fluxes declined rapidly in both processes and steady state conditions were reached after a few hours of operation. For a comparison purposes, the average permeate flux from each process for steady state conditions achieved in each 5-day cycle are presented in Fig. 4. Figure 4 shows that during the first cycle of operation, the average permeate fluxes from both processes were almost the same as the EC step was turned off during the operation of the first cycle.

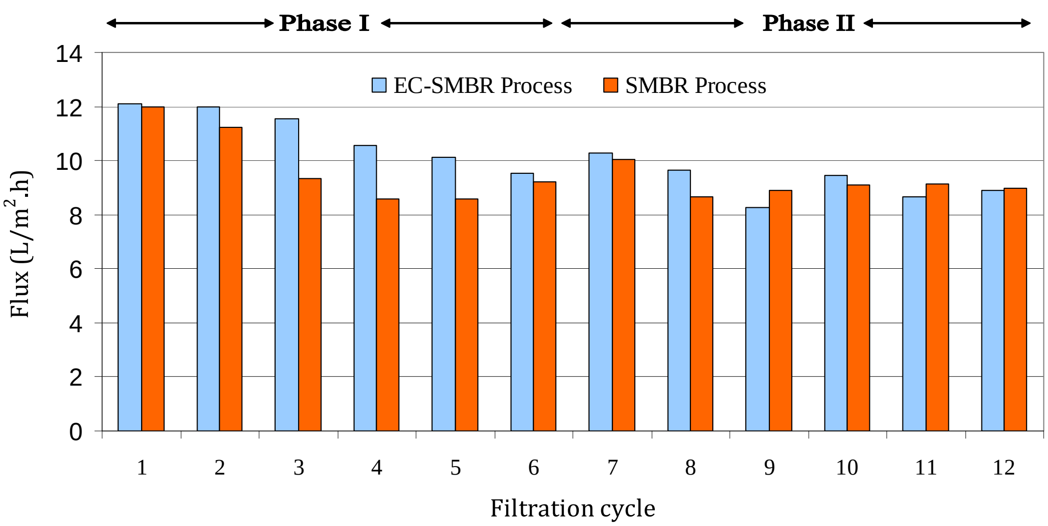

Figure 4. Average permeate fluxes from EC-SMBR and SMBR processes during the two experimental phases

Starting from the second cycle, a small difference was observed between the permeate fluxes from the two processes; the figure shows that the average permeate flux from EC-SMBR process was higher than the average permeate flux from SMBR system alone in Phase I. This may be attributed to the agglomerated particles produced as a result of applying an electrocoagulation step.

The improvement in permeate flux was more obvious in cycles 3 to 5 of Phase I. This suggests that the activated sludge should be coagulated before starting the operation of the membrane module to prevent the chance of the small particles to go inside the membrane pores and clog them. This conclusion was recommended by another study conducted on treating synthetic municipal wastewater by submerged membrane electro-bioreactor (Bani-Melhem and Elektorowicz, 2010).

The improvement in membrane flux in EC-SMBR process was associated with an increase in microbial flocs size which was measured after each experimental cycle (Fig. 5). No significant variations were observed in the microbial flocs size of the SMBR process during Phase I; however, a small but measurable increase was observed in Phase II. On the other hand, beginning from cycle-2, the microbial floc size in the EC-SMBR process increased considerably with time, with the mean floc size reaching 208 μm at the end of Phase II.

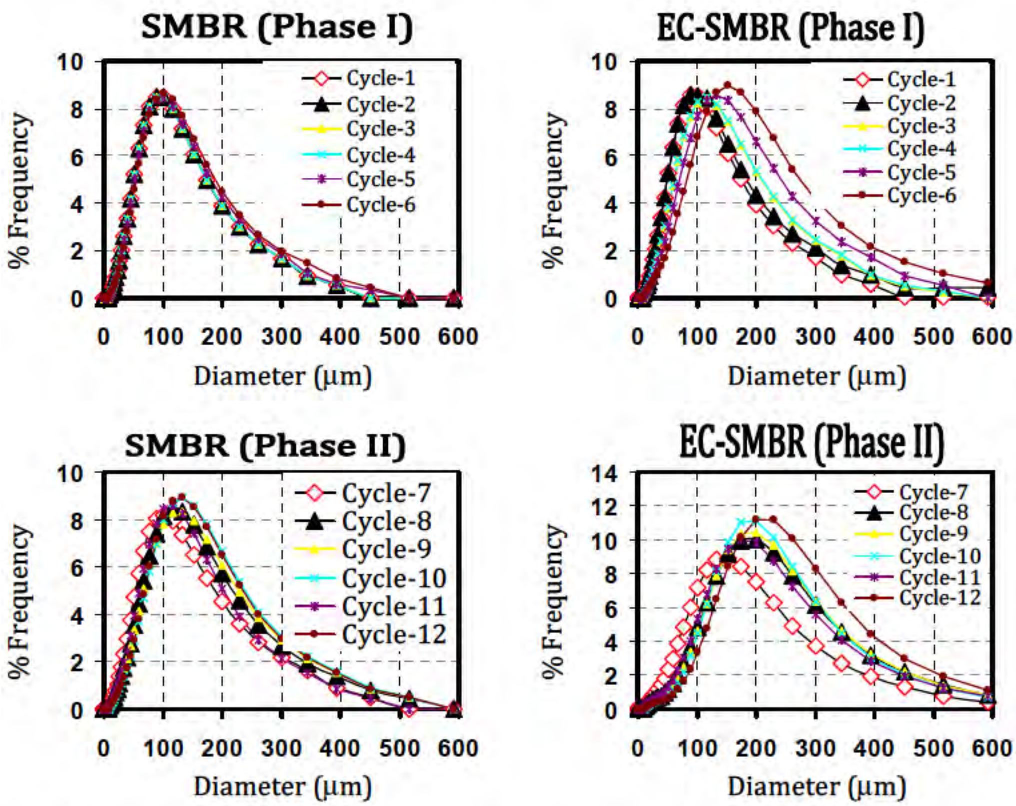

Figure 5. Variations of the particles size distribution of sludge suspensions in SMBR and EC-SMBR processes during twelve cycles of the two experimental phases

However, it seems that the increase in the particle size did not play a significant role in decreasing the membrane fouling in Phase II as the improvement in membrane permeability was not observed in all sequential cycles although the microbial flocs size was greater in EC-SMBR process. For example, the permeate flux from EC-SMBR process alone was relatively better in cycles 7, 8 and 10 of Phase II, but the SMBR system achieved relatively better performance in cycles 9 and 11 and no significant difference as observed in cycle 12. This trend in membrane permeability in EC-SMBR process in Phase II may be due to two main factors: (1) operation of the bioreactor at complete sludge retention time (SRT) might lead to accumulation of inorganic matter on the membrane surface, and (2) the large increase in anionic surfactants during Phase II could contribute in reducing the membrane filtration performance in SMBR processes in general. The negative impact of anionic surfactants concentration on membrane flux was observed in a study conducted by Dhouib *et al*. (2005).

It is also plausible that the increase in aluminum dosage in the bioreactor of EC-SMBR process might play a role in increasing membrane fouling. Previous research has reported that fouling was alleviated only up to an intermediate value of aluminum dosage beyond which higher coagulant dosages actually increased fouling (Lee *et al*., 2001; Ben Sasson and Adin, 2010). Although not the objective of this study, optimization of applied EC voltage together with aluminum (or other anode) dosage is an important area of further research.

CONCLUSIONS

The performance of an electrocoagulation unit operated in series with a submerged membrane bioreactor system fed with real grey water was explored in this study in terms of the potential to decrease membrane fouling and enhance pollutant removals. The operation of the EC-SMBR process was compared with the operation of a conventional submerged membrane bioreactor process. Both processes were operated at constant transmembrane pressure and complete sludge retention time.

The comparison was achieved in two sequential operational phases in which two different voltage gradients were investigated in EC step, namely 0.64 and 1.26 V/cm. The results of this study demonstrated the following conclusions:

- An adequate voltage gradient should be applied in EC step to achieve a significant increase in pollutant removals. In this study, a notable improvement in permeate quality was observed at 1.26 V/cm rather than 0.64 V/cm.
- The production of aluminum should be controlled and wasted from the bioreactor to prevent the accumulation of inorganic materials on the membrane which might negatively affect membrane permeability.
- The impact of anionic surfactants concentration on membrane flux should be explored in the future work.

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