

# Submerged Membrane Electro-Bioreactor (SMEBR) Reduces Membrane Fouling and Achieves Phosphorus Removal

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## ABSTRACT

The occurrence of membrane fouling is still the major factor hindering the wide-spread use of membrane bioreactor (MBR) technology. Furthermore, enhanced biological phosphorous removal would be limited in MBRs applications. The main objective of this study was to develop the Submerged Membrane Electro-Bioreactor (SMEBR), for decreasing the fouling rate and increasing the performance in term of phosphorus removal. Combining biological, membrane and electrochemical processes in one operation SMEBR unit has achieved 52 % reduction in membrane fouling and more than 98% efficiency of PO<sub>4</sub>-P removal.

**KEYWORDS:** Submerged Membrane Electro-bioreactor, Membrane fouling, Electrocoagulation, Phosphorous removal

## INTRODUCTION

Membrane bioreactor (MBR) is becoming an established technology in treatment of municipal wastewater, with a number of installations in the biological nutrient removal exceeding capacity of 50 ML/d (Canham 2009). Initially the membrane module was utilized outside the reactor in an external configuration. Further developments of the process placed the membrane directly in the activated sludge reactor, thus integrating the biological process with solids separation (Yamamoto et al., 1989). This configuration, called a submerged membrane bioreactor (SMBR) was used to lower the energy costs in smaller installations (Ueda et al., 1997; Yamamoto et al., 1989). Today, most full scale larger MBR are conventionally designed as a system of a separate bioreactor tank with mixed liquor flowing to separate tank housing the submerged membranes (Canham 2009).

Although SMBRs solved many problems associated in activated sludge processes (ASP), fouling of the membrane is the major factor in reducing the wide-spread use of the process (Li and Wang, 2006; Le-Clech et al., 2006; Chang et al., 2002; Judd, 2005). Furthermore, because SMBRs are often operated with minimum sludge removal, holding

high concentrations of the sludge by maintaining long sludge retention time (SRT), enhanced biological phosphorous removal (EBPR) would be limited in MBR applications (Adam et al., 2002).

Many studies have been conducted to solve the problem of membrane fouling in SMBR applications. Generally, most of the conducted studies to reduce SMBR fouling can be grouped by three distinct approaches: cleaning the membrane unit, optimizing the operating parameters and improving the wastewater characteristics (Bani-Melhem, 2008).

Cleaning the membrane is achieved physically by backwashing the membrane module using the permeation flux or back flushing using a high flow rate stream of air. In the long run, chemical cleaning is required for the membranes to recover its permeability. Optimization the operating parameters includes the selection of the best operating conditions in terms of aeration intensity, sludge retention time (SRT), hydraulic retention time (HRT) or mixed liquor suspended solid (MLSS) concentration in the bioreactor to minimize the fouling on the membrane. Improving the characteristics of the treated wastewater has been proven an effective approach in reducing the fouling in submerged membrane bioreactors applications. This approach includes the addition of chemical coagulants such as alum and iron salts to increase the floc size of the MLSS solution (Song et al., 2008; Wu and Huang, 2008; Wu et al., 2006; Lee et al., 2001) or the addition of adsorptive materials like high concentration of powdered activated carbon (Guo et al., 2008; Hu and Stuckey, 2007; Munz et al., 2007; Lee et al., 2006; Seo et al., 2004) and zeolite (Lee et al., 2001).

In terms of phosphorus removal, many studies have been conducted to enhance the phosphorus removal in SMBR applications in parallel with reducing the fouling rate. These studies include modifying green bio-flocculant (Ngo and Guo, 2009), the addition of sponge (Ngo et al., 2008) and coagulant addition (Song et al., 2008).

All of the techniques have flaws such as significant increase of the operating costs; potential for membrane damage (Le-Clech et al., 2006); side effects of the generated by-products and/or increasing the volume of sludge in the reactor (Clark and Stephenson, 1998).

Recently a new technology was developed at Concordia University, Montreal, called Submerged Membrane Electrokinetic Bioreactor or SMEBR (US Patent, 2008). The new technology was designed based on applying a direct current (DC) field between immersed circular electrodes around immersed membrane filtration module. The treatment of wastewater with the SMEBR system involves simultaneous application of biodegradation, electro-coagulation and filtration through membrane. Detailed description of the design constraints and criteria of the new developed method was reported by Bani-Melhem (2008).

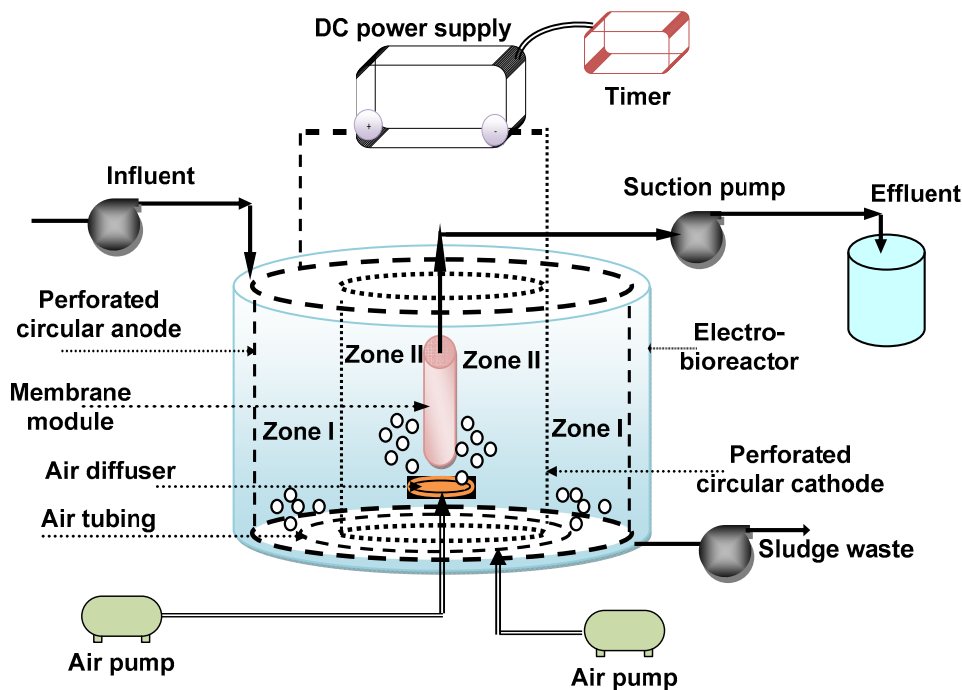
This application of the SMEBR to treatment of wastewater while reducing membrane fouling achieved by changing the operational conditions and characteristics of wastewater, while improving phosphorous removal will be presented.

**METHODOLOGY**

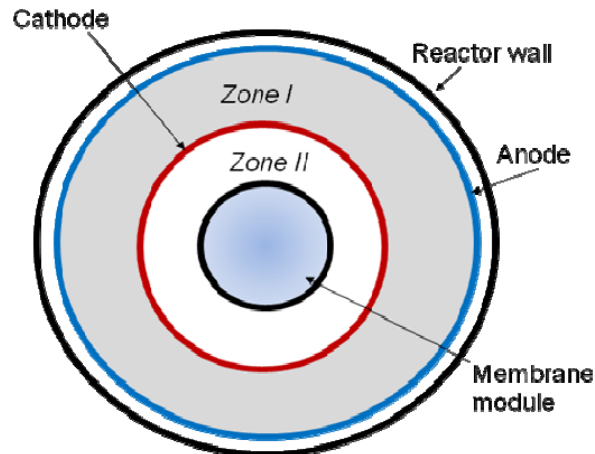
**Experimental Setup**

Laboratory scale experimental set-up comprised an electro-bioreactor with a working volume of 13.4 L, a membrane module, an aeration system, and a direct current (DC) supply system – Fig. 1. Two cylindrical electrodes (anode and cathode) were used. The anode was made of perforated aluminum sheet while the cathode was made of iron mesh. Electrodes were connected to a digital external DC power supply (TES 6230). Timer model 5500 (Control Company, USA) was connected with the DC power supply to regulate the operating mode of an interrupted supplying DC field due to presence of microbial culture. Submerged hollow fiber membrane module ZeeWeed-1 (GE / Zenon Membrane Solutions, Canada) was fixed vertically in the center of the electro-bioreactor and it separated treated effluent from the mixed liquor.

The design of the SMEBR system divides the reactor into two zones (Fig. 2): Zone I (electro-bioreactor) extends from the external wall of the reactor to the cathode and Zone II is located between the cathode and the membrane module. Generally, Zone I is dominated by the processes of biodegradation and electro-coagulation while Zone II is responsible for further biodegradation and membrane filtration.



**Figure 1. Schematic diagram of the experimental setup.**



**Figure 2. Top view of the submerged membrane electro-bioreactor (SMEBR)**

The system of aeration in the submerged membrane electro-bioreactor consisted of three parts. The first part is a porous air diffuser was installed just below the membrane module in the bottom center of the electro-bioreactor in the Zone II. It supplied air continuously with the help of an air pump to maintain the required dissolved oxygen (DO) level above 5 mg O<sub>2</sub>/L. The supplied air also agitated the sludge and was supposed to reduce membrane fouling. The second part of the aeration system was a perforated air tube which was used for keeping biomass in suspension and to supply air to help to maintain the DO > 5 mg/L in Zone I. The aeration system was also used to pumping air continuously (third part) through the membrane module as designed by the manufacturer to reduce fouling and cake formation

### **Feed Wastewater**

The synthetic wastewater used in the experiments was made with tap water and consisted of (all in mg/L): Glucose (310), Peptone (252), Yeast extract (300), (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (200), KH<sub>2</sub>PO<sub>4</sub> (37), MgSO<sub>4</sub>·7H<sub>2</sub>O (40), MnSO<sub>4</sub>·H<sub>2</sub>O (4.5), FeCl<sub>3</sub>·6H<sub>2</sub>O (0.4), CaCl<sub>2</sub>·2H<sub>2</sub>O (4), KCl (25), and NaHCO<sub>3</sub> (25). About 40 L of synthetic wastewater was prepared once per week (av. influent: COD = 350 mg/L, PO<sub>4</sub>-P = 26 mg/L, NH<sub>3</sub>-N = 45 mg/L). The sludge inoculums were taken from the secondary clarifier in the municipal wastewater treatment plant in the City of Saint-Hyacinthe (QC, Canada). The biomass was acclimated to the synthetic wastewater for two months in order to achieve stable conditions prior to the membrane filtration experiments.

### **Experimental Procedure**

The reactor operated continuously for a period of 66 days without wasting of biosolids (an infinite SRT) while being continuously fed with the synthetic wastewater. The aeration was continuous in both zones. The process was operated at a constant trans-membrane pressure, that is the permeate flux gradually decreased with time due to the fouling phenomena. Moreover, no backwashing of the membrane module was performed during the operation although the membrane module was periodically taken out of the electro-bioreactor, when the permeate flux significantly declined and externally

washed with distilled water for a few minutes to remove the attached sludge cake particles over the membrane surface.

There were four subsequent research stages:

- The Stage I: from day 1 to day 33; the reactor worked simulating conventional submerged membrane bioreactor (SMBR). No current was applied, however, metallic electrodes were already placed dividing the bioreactor in the same two zones as in subsequent stages;
- The Stage II: day 33 to day 40; first stage of SMEBR investigation;
- The Stage III: day 40 to day 57;
- The Stage IV: day 57 to day 66.

During Stages II, III and IV, the SMEBR was connected to the power supply with two different operational modes (15' ON/45' OFF and 15' ON/105' OFF). Table 1 shows the different operating conditions. A constant voltage gradient of 1 V/cm was applied in stages II, III and IV based on preliminary studies (Bani-Melhem, 2008).

The membrane was cleaned with distilled water wash (no back-washing) after Stage I; however, chemical cleaning was applied after Stage II as distilled water surface washing was found inadequate. The membrane module was removed from the reactor and immersed in tap water for 2 hours, and subsequently in 5% NaOCl solution for 8 hours. It was finally rinsed with tap water. This procedure enabled to re-establish most of the membrane's permeability. Similar procedure was done by Meng et al. (2006).

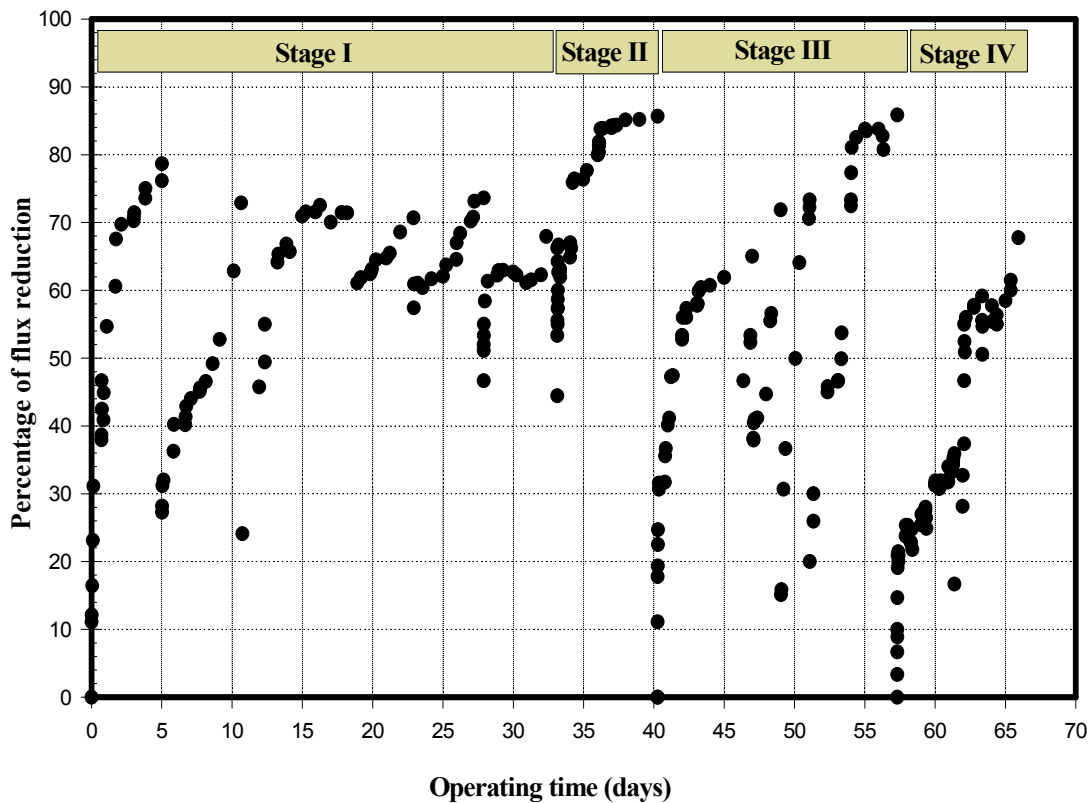
**Table 1. Experimental conditions**

Items	Stage I	Stage II	Stage III	Stage IV
Operation time (days)	33	7	17	9
SRT (days)	≈200	≈200	≈200	≈200
DO in bioreactor (mg/L)	5-8	5-8	5-8	5-8
DC (V/cm)	0	1	1	1
DC exposure time (minutes)	0	15 ON / 45 OFF	15 ON / 45 OFF	15 ON / 105 OFF
Strategy of applying DC field	Reference only. No DC current (similar to conventional MBR).	Simultaneously with the operation of the membrane	Applied before operating the membrane	Applied before operating the membrane
Influent temperature (°C)	19-21	19-21	19-21	19-21

**RESULTS**

Since the operational period was divided into four stages according to the experimental conditions mentioned in Table 1, similar measurements and analyses were performed in each stage. The results of the permeate flux reduction changed during the entire experiment and during the first five days of each stage - Fig. 3 and Fig. 4. The SMEBR system performed best during Stage IV in terms of membrane filtration. In Stage IV the flux reduction was 38 % after 5 days of operation without membrane cleaning, which corresponds to 52% less fouling than in a conventional submerged membrane bioreactor (simulated by Stage I).

The SMEBR system achieved also excellent removal of phosphorous reaching 98.3% (Fig. 5).



**Figure 3. Percentage reduction in permeate flux (*PRPF*) in submerged membrane bioreactor operation (Stage I) and SMEBR system operation (Stages II, III and IV)**

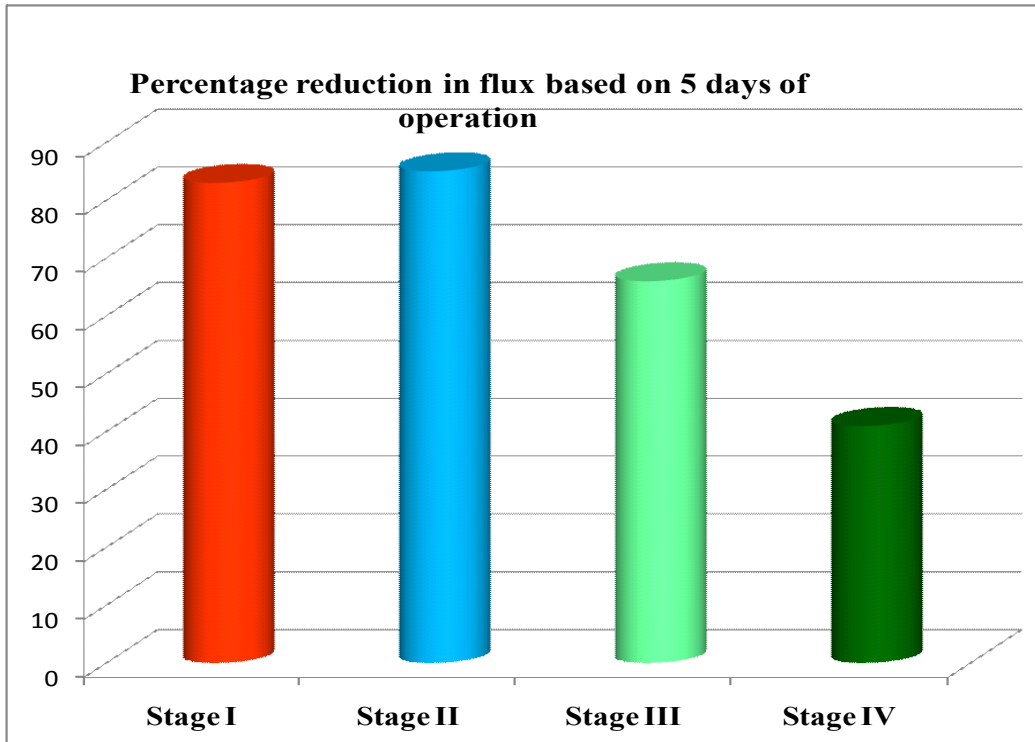


Figure 4. Improvement of performance due to electrokinetically fouling control.

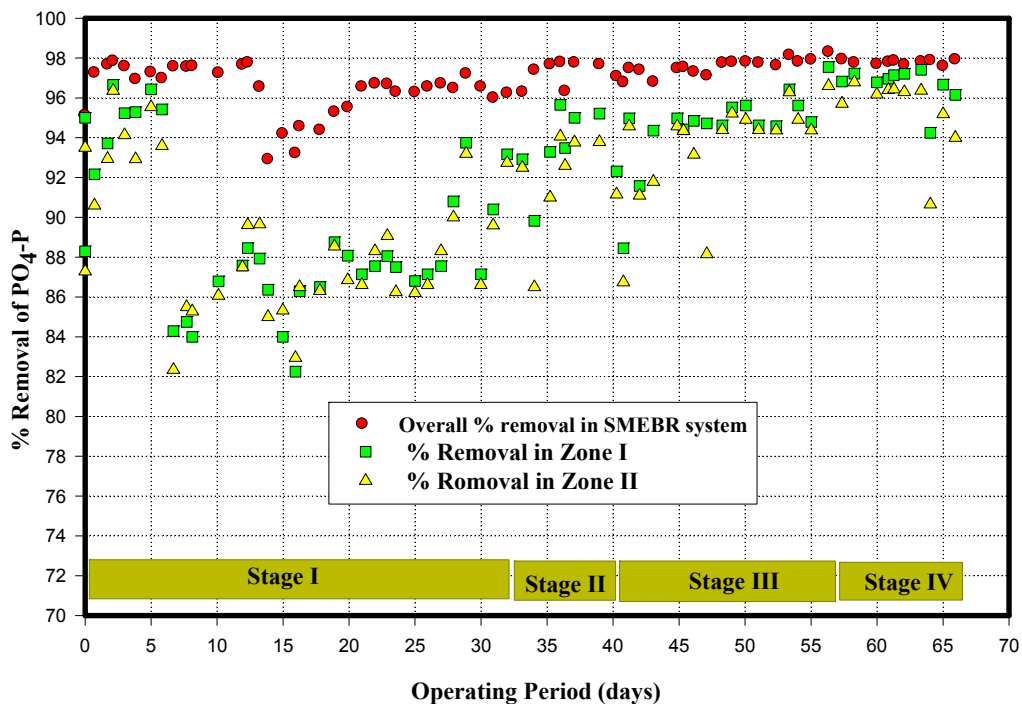


Figure 5. Phosphorus removal in SMEBR system: Stage I was without electrical field (i.e. operated as a conventional MBR)

## DISCUSSION

### Impact of the SMEBR Operation on Membrane Filtration Performance

Since the SMEBR system was operated under a mode of constant trans-membrane pressure, the decrease of permeate flux ( $J$ ) with time showed the membrane fouling changes. To assess the fouling behavior in the SMEBR system during the four stages, the concept of percentage reduction in permeates flux ( $PRPF$ ) was used (shown in Fig.3):

$$PRPF = \left( \frac{J_i - J}{J_i} \right) \times 100\% \quad (1)$$

where  $J_i$  is the initial permeate flux ( $2.72 \times 10^{-6} \text{ m}^3/\text{m}^2 \cdot \text{s}$ ) and  $J$  is the permeate flux at any time.

During Stage I, with no input of DC power, the permeate flux decreased very quickly due to fouling. After 5 days of operation of Stage I, the  $PRPF$  was 80%. To enhance membrane permeability, the membrane was washed with distilled water for few minutes outside of the reactor when the ( $J/J_i$ ) ratio reached 0.2 - 0.3 on days 5, 11, 19, 23 and 28.

The objective of operation Stage II was to investigate the performance of the SMEBR system when the DC is applied to the MLSS solution in conjunction with the operation of the membrane module. At the end of Stage I, the membrane was removed for physical cleaning by distilled water for a few minutes outside the electro-bioreactor, and subsequently immersed in the electro-bioreactor. Immediately after applying a DC field with a mode of operation of 15' ON/ 45' OFF, the flux declined faster than in Stage I ( $PRPF = 82\%$  after 5 days of operation). It was hypothesized that: i) the washing by physical cleaning before operating Stage II was not sufficient to remove all membrane deposits from Stage I; ii) the electro-coagulation process did not have enough time to form aggregates, which would directly affect the membrane filtration process. The expectation was that electrokinetic phenomena would form the flocs which in turn would limit the transport of fine particles towards membranes and their deposition, thus limit fouling.

Accordingly, physical and chemical washing of membrane was done before the beginning of Stages III and IV. To prepare the MLSS solution for coagulation, the SMEBR system was operated with the DC field but without membrane module during the first hour of the Stage III; subsequently, the membrane module was immersed in the electro-bioreactor. At this stage, the influence of the electro-coagulation process is clearly reflected in the membrane filtration process, since the permeate flux declined much lower than observed in Stages I and II. During Stage III, the reduction in membrane flux was at around 63% after 5 days of operation.

In Stage IV, the mode of operation of the DC was modified to 15 minutes ON / 105 minutes OFF. The results from this stage showed that the  $PRPF$  was only 38% after 5 days of the continuous operation without any kind of backwashing and external chemical washing of the membrane module. By comparing the first five days of operation of each stage, the SMEBR system performed best during Stage IV in terms of membrane filtration when compared with any other stage (Fig. 4).



### Phosphorous Removal Performance

The main reactions of electrocoagulation with aluminum used as the anode (Koby et al., 2006) are:

- At the anode:



- At the cathode:



- In the solution:



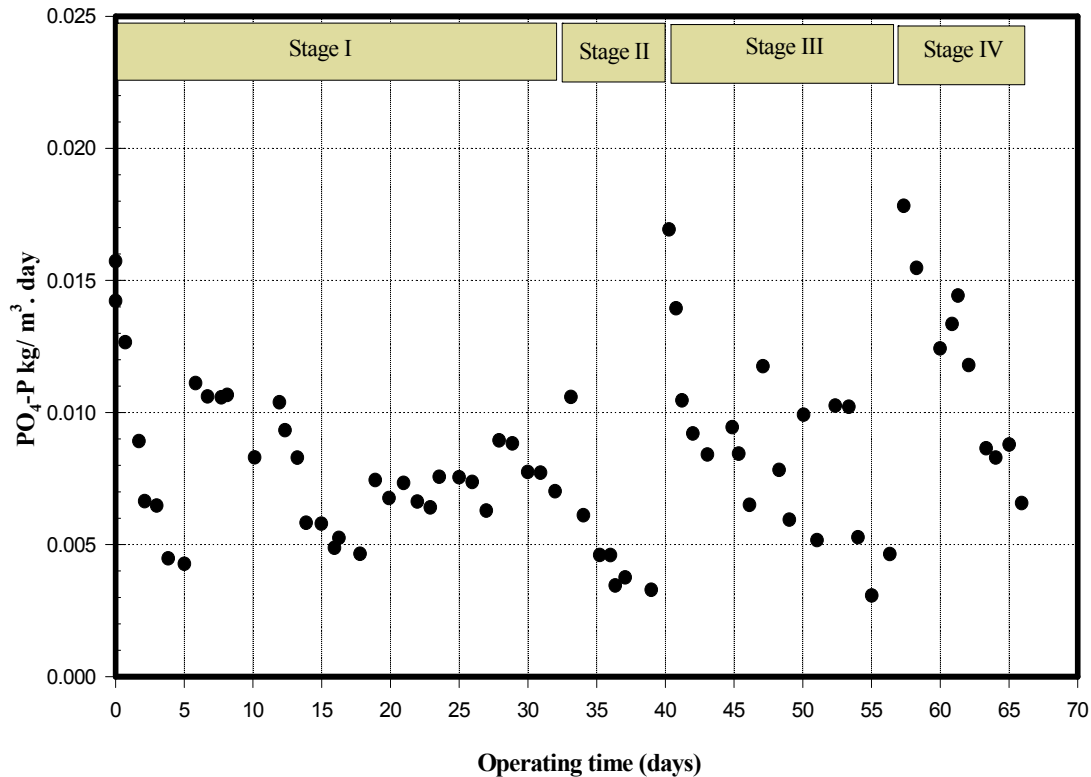
The amorphous  $\text{Al} (\text{OH})_3(\text{s})$  formed is characterized by “sweep flocs” that have large surface areas which are beneficial for the rapid adsorption of soluble organic compounds and entrapping colloidal particles (Koby et al., 2006). These flocs are removed easily from aqueous medium by sedimentation. On the other hand, aluminum ions ( $\text{Al}^{3+}$ ) react with phosphorous ions to form  $\text{AlPO}_4$  which precipitates according to the following reaction:



Fig. 5 shows an excellent phosphorous removal due to electrocoagulation in Stages II – IV. The phosphorus removals varied from the minimum value of 96% in Stage II to a maximum of 98.3 % in Stage IV.

As the SMEBR system operated at constant pressure mode, the hydraulic retention time varied/fluctuated with the operation due to decline of permeate flux due to fouling. The HRT affects the volumetric loading to the SMEBR system as shown in Fig. 6. Consequently,  $\text{PO}_4\text{-P}$  volumetric loading to the system changed continuously as shown in Figure 6. These trends had impact on the biological removal efficiencies.

Starting from Stage II (day 33 to day 40), after applying a DC field into the MLSS solution, an increase in the  $\text{PO}_4\text{-P}$  removal was observed in the SMEBR’s zones. This can be understood from the electrokinetic phenomenon that occurred in Zone I. The metal ions ( $\text{Al}^{3+}$ ) began to appear in the MLSS solution forming aluminum hydroxide which functions as sweep flocs. These sweep flocs have large surface areas which are best suited for P removal. The improvement in phosphorus uptake continuing during Stages III and IV was associated with the further building of aggregates.

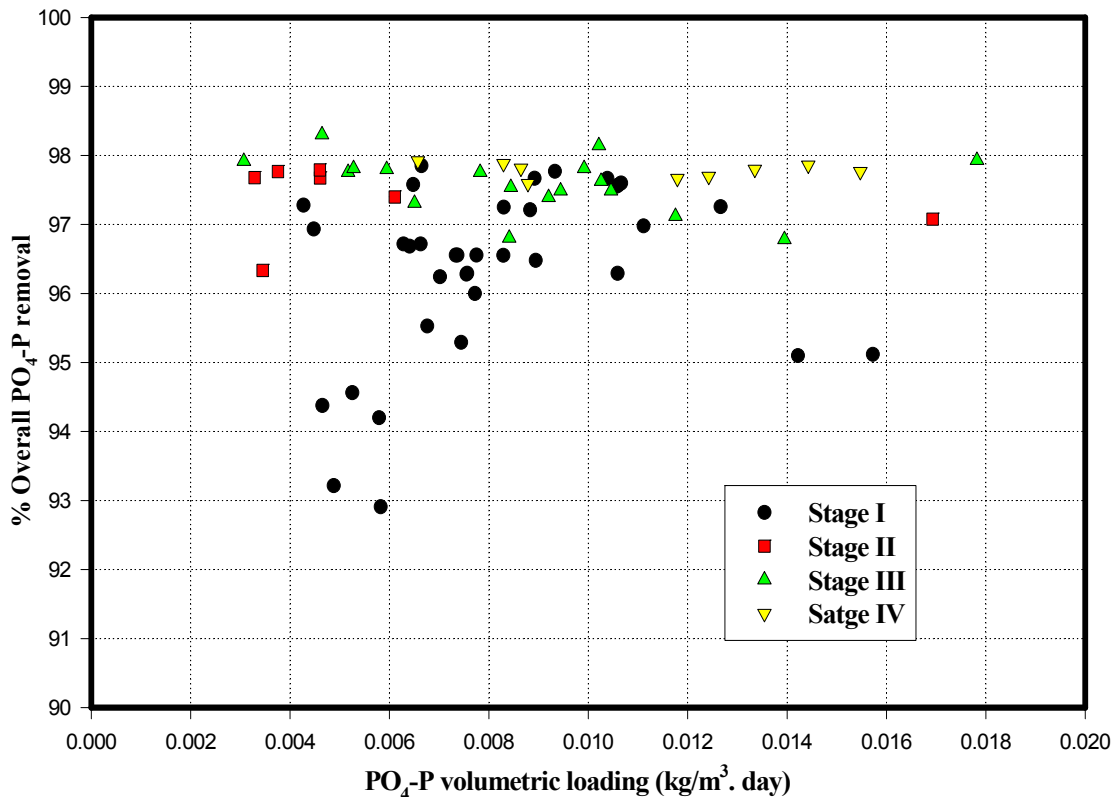


**Figure 6. Changes in phosphorus volumetric loading during the operation of SMEBR system: Stage I was without electrical field (i.e. operated as a conventional MBR)**

These results demonstrated that during the first stage of operation, where no DC was applied, the concentrations of volumetric loading in both zones of the electro-bioreactor supernatants were similar to each other; while after application of DC, little differences were observed between the two zones. The concentration of PO<sub>4</sub>-P in Zone I was on average less than 2-4% the concentration of PO<sub>4</sub>-P in Zone II. This is attributed to the coagulation phenomenon that occurred in Zone I. As the aluminum ions (Al<sup>3+</sup>) are produced in Zone I, as a result of the electrochemical process, the phosphorous molecules reacted with aluminum ions to produce AlPO<sub>4</sub> and precipitated as shown in Equation 5. Alternately, the aluminum ions (Al<sup>3+</sup>) react with the hydroxyl ions (OH<sup>-</sup>) produced at the cathode to form Al(OH)<sub>3</sub> flocs in Zone I. The phosphorous molecule will get a better chance to be adsorbed onto these flocs and precipitated out to the bottom of the reactor. In Zone II, where theoretically, no electro-coagulation process took place, there was less chances for the available phosphorous molecules to react with aluminum ions; therefore, the removal efficiency of phosphorus in Zone II was less than the removal efficiency of phosphorus in Zone I.

To analyze the impact of changes of volumetric loading on the overall performance of the SMEBR system, the overall removal of ortho-phosphorous were plotted versus the variations in the PO<sub>4</sub>-P volumetric loading as shown in Figure 7. Figure 7 demonstrates

that the % removal in PO<sub>4</sub>-P was always greater than 96 % during the Stages II, III and IV in which the electrocoagulation took place. The only decrease in PO<sub>4</sub>-P removal was observed in Stage I which might be due to the fluctuation in the HRT. However, because electrocoagulation, the designed SMEBR system can supply a continuous production of flocs which might be able to deal with the sudden increase in the volumetric loading of PO<sub>4</sub>-P. This means that the designed SMEBR system can deal with sudden shock in volumetric loading which considers another advantage of the designed SMEBR system.



**Figure 7. Variations of overall removal efficiency of phosphorus in SMEBR system with volumetric loading: Stage I was without electrical field (i.e. operated as a conventional MBR)**

**CONCLUSIONS**

Submerged membrane electrokinetics bioreactor SMEBR was fed synthetic wastewater to test its ability to decrease membrane fouling and enhance phosphorous removal. The operation of the SMEBR system was compared with the operation of conventional membrane bioreactor at high SRT. The overall PO<sub>4</sub>-P removal was greater than 98% and appeared independent of the volumetric load. The SMEBR has significantly reduced membrane fouling demonstrating 52% less fouling (at constant permeate flux operation) than the conventional MBR system. The SMEBR system developed opened another avenue of expanding the applicability of membrane processes to treatment of wastewater.

## ACKNOWLEDGMENTS

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