



## Removal of phosphate and suspended solids by electrocoagulation coupled with woven fabric membrane at pilot plant

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### ABSTRACT

The objective of this study was to develop a rapid, economical, and eco-friendly process for the efficient removal of phosphate and suspended solids (SS) from the wastewater in the continuous mode at pilot plant scale. In this method, electrocoagulator (EC) was coupled with woven fabric membrane and the effluent was removed directly from the reactor through the membrane thus omitting the settling stage. The experiment was carried out for four months by maintaining an optimal current density of 0.8 mA/cm<sup>2</sup> in the EC reactor. No chemical injection was made at any stage of the investigations and the membrane was operated at very high flux rates ranging from 25 to 75 LMH. The phosphate and SS removal efficiencies of the developed hybrid method were around 98 and 99%, respectively, while the removal efficiencies of these contaminants were between 78 and 61%, respectively, by providing only EC treatment. The final effluent phosphate and SS contents were less than 0.20 and 0.35 mg/L, respectively, in the treated wastewater. The removal rates were identical at all flux rates and the maximum increase in transmembrane pressure was 10 kPa at 75 LMH. SEM images showed that woven fabric membrane had excellent regeneration ability. It had high robustness, generated good quality effluent, and only needed physical cleaning.

*Keywords:* Phosphate; Suspended solids; Electrocoagulation; Woven fabric membrane

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### 1. Introduction

The main purposes of wastewater treatment are to protect the environment and conserve the fresh water resources. The quality of treated water before its disposal to the environment is closely examined to ensure the strict policies established by various environmental protection agencies. Eutrophication is one

of the major challenges encountered in the monitoring of environmental water sources. This phenomenon is generally due to the phosphorus present in the wastewater and when such water is discharged into the lakes or rivers, results in the enhanced growth of algae and other biomass. It is very dangerous for the ecosystem, and the decomposition of such large amounts of biomass significantly reduces the oxygen level in the water body, causing the reduction in fish

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and other water species [1,2]. In Europe, the discharge of municipal wastewater to sensitive watercourse is only permitted, if it has phosphorus contents between 1 and 2 mg/L [3]. Even, very low phosphorus concentrations (100 µg/L) provide sufficient nutrition enriched atmosphere to microbes in the lakes [4]. Similarly, high concentrations of suspended solids (SS) are a major threat to aquatic life and stream health. The presence of SS in the treated water blocks the light reaching the submerged vegetation, thus affecting their photosynthesis process. Consequently, the plants are decomposed leading towards the severe shortage of oxygen and destroying the whole ecology. SS also block the fish gills, reduce their growth rates and have adverse effects on larval developments in the water stream [5]. High SS contents in the effluent water also limit its applications for industrial usages as these can clog and scour pipes and equipment.

Considering these destructive environmental impacts of phosphorus and SS, various physical, chemical, and biological methods have been used for their removal from wastewater like membrane separation, electrodialysis, coagulation, adsorption, precipitation, and so on [6–9]. But the problem is that the chemical methods are expensive, health hazardous, and also not very effective at low contaminant concentrations. The other disadvantages associated with these processes are that they produce a large amount of sludge and also increase the concentration of total dissolved solids in the wastewater during the treatment process [10]. The biological processes are very slow and also require high strength wastewater for phosphorous removal, which is not possible until the addition of supplementary carbon [1,4,11]. On the other hand, membrane fouling is a major obstacle in using the membrane technology for wastewater treatment as it increases the operating cost. The fine organic and inorganic suspended particles not only accumulate on the membrane surface, but also deposit inside the membrane pores, thus causing severe pore-blocking. The cleaning of such fouled membranes is another problem as it involves very toxic and expensive reagents and also reduces the permeability by aging of the membrane [12].

Therefore, there is a dire need to develop an economical and eco-friendly process that can efficiently remove phosphorous and SS from the wastewater. The research works reported in the last few years have shown that electrocoagulation (EC) process has the potential to be an alternative method for the removal of these contaminants [13,14]. During the EC treatment, no chemical compounds are injected for the wastewater treatment, thus eliminating the health risks and less generation of chemical sludge [15]. In

the EC process, the reactive iron or aluminum electrodes are used which electrostatically generate the respective metal ions in the aqueous solution. These metallic ions attract the negatively charged colloidal particles from the wastewater and agglomerate the fine suspended particles into large flocs which eventually settle down under gravity in the settling tank and later removed from the system. Similarly, the release of metal ions from the anodes precipitates the phosphorus as metallic phosphates to help its removal from the wastewater [13–16].

Most of this EC published data are based on the laboratory studies and less information is available about plant scale experiments. However, there are some concerns like time and cost of EC method, which need to be addressed prior to its use on a commercial scale. The EC process involves the settling phase and generally operates in the batch mode in which appropriate time is required to the precipitated material for its settling and removal from the system. Furthermore, high current density is normally applied to remove the phosphorus down to 2 mg/L, which eventually decrease the electrode age and increase the operational cost. The objective of this research work was to modify the EC process in order to increase its phosphorous and SS removal efficiencies by taking into account the above mentioned challenges. In our modified EC system, EC was (operated at low current densities) coupled with submerged woven fabric membrane and operated in the continuous sequence at pilot plant. The treated water was directly removed from the reactor by membrane, thus, omitting the need of the settling chamber. It was expected that the use of this hybrid process will increase the recovery of phosphate and fine suspended particles and the continuous operation of wastewater treatment at plant scale could be achieved.

## 2. Materials and methods

### 2.1. Operation of modified EC reactor

The pilot plant for these studies was installed at Ilsan wastewater treatment plant, Goyang, South Korea. The operational period of the experiment was four months. The wastewater from the secondary clarifier of this treatment plant was used as influent and pumped to the storage tank before its feeding to the EC reactor. The average composition of feed water is listed in Table 1. The dimensions of reaction tank were  $W$  0.8 m  $\times$   $L$  1.2 m  $\times$   $H$  1.0 m, having total volume 960 L and workable volume 800 L. A level controller was fixed on the influent pump to maintain the workable water level in the reactor. EC was equipped with

Table 1  
Characteristics of feed water used in the pilot plant studies

Constituents	Average concentration	Variation
pH	7.0	±0.20
Alkalinity (mg/L as CaCO <sub>3</sub> )	120	±18
SS (mg/L)	65	±7
PO <sub>4</sub> -P (mg/L)	7.5	±1
COD (mg/L)	55	±5
Conductivity (µs/cm)	810	±20

two parallel iron plates, which worked as electrodes and installed at 2 cm distance from each other. The dimensions of iron plate were  $T$  2 mm  $\times$   $L$  0.8 m  $\times$   $H$  0.8 m and its active surface area was 1.28 m<sup>2</sup>. The experiment was performed in a continuous mode and the electric current was provided via the iron electrode into the EC reactor, which liberated iron into the liquid medium for the precipitation of contaminants. The current density (calculated as the current supplied per unit area of iron plate) was optimized and maintained at 0.8 mA/cm<sup>2</sup>. Two woven fabric membrane modules were immersed horizontally in the reactor on the plastic baffles. Air diffusers were installed about 50 cm above the reactor basin to enhance the mass transfer from bulk to the electrode, and also to continuously clean the membrane surface. The height of air diffuser was purposely designed at a high level so that the precipitates could be settled on the bottom of the reactor, from where precipitates were periodically discharged outside through a precipitate discharge line. The whole process was automatically controlled

through the control panel. The schematic diagram of the pilot plant is shown in Fig. 1.

The treated water was directly removed from the EC reactor through the three-dimensional woven fabric membranes and discharged into the effluent stream; this was regarded as Eff-1. To check the efficiency of the modified process (EC coupled with woven membranes), the same amount of treated water was also discharged continuously from the EC reactor through the settling chamber without using the membranes and termed as Eff-2 (control sample).

## 2.2. Characteristics and operation of woven fabric membrane

Two identical membrane modules were employed for filtration; each module was made up by bunching five segments of woven fibers, each segment containing 80 cm long 100 pieces of fiber. The total surface area of membrane was 1.8 m<sup>2</sup>. The treated water was removed by using a centrifugal pump at normal atmospheric pressure. The filtration was conducted at three different flux rates, i.e. 25, 50, and 75 LMH for 60, 30, and 30 d, respectively, and any variation in the trans-membrane pressure (TMP) during the experiment was noted through the attached gage. The instantaneous flow and total treated volume were noted by installing the respective flow meters. The flux rate was calculated by using the following formula.

$$\text{Membrane flux} = \frac{\text{Volume (L)}}{\text{Membrane area (m}^2\text{)} \times \text{Time (h)}} = \text{LMH}$$

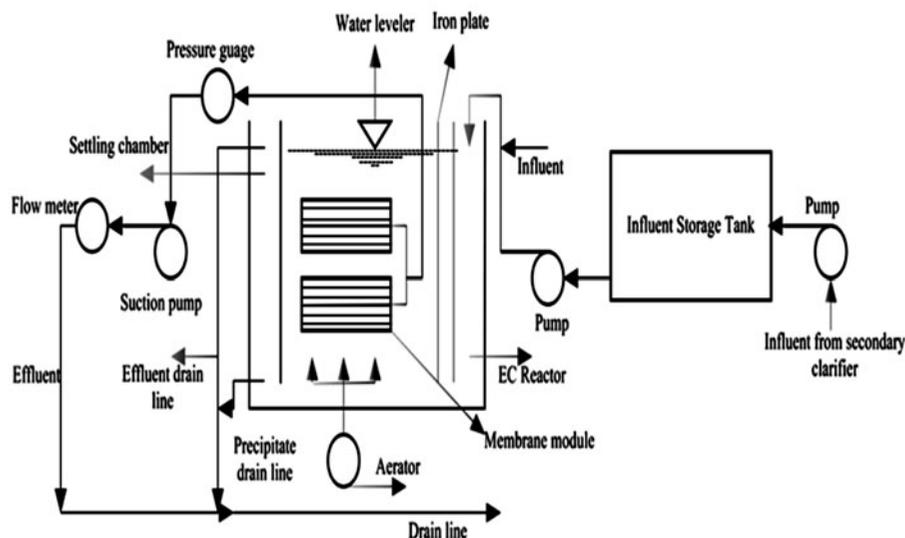


Fig. 1. A schematic diagram of pilot plant reactor.

The membrane was operated intermittently in on/off cycles of 6 and 2 min, respectively. During the process, membrane was only cleaned physically by brushing and spraying the tap water on it and no chemical cleaning was performed.

### 2.3. Analytical determinations

The influent and effluent samples were regularly analyzed for the determination of different constituents to monitor the performance of pilot plant. Total phosphate contents ( $\text{PO}_4\text{-P}$ ) were noted with a Hach spectrophotometer (DR/4000) using molybdovanadate method. Total iron and COD were measured by employing Hach program 2165 and 2710, respectively. pH determination was made with a pH meter (Model 3510, Jenway Ltd., UK), while SS and alkalinity measurements were performed as described in the standard methods [17].

### 2.4. Scanning electron microscope (SEM) measurement

The membrane surface was monitored by using SEM (Hitachi S-4800, Japan). The pieces of used and physically cleaned membrane were cut from the membrane module and fixed with 2% glutaraldehyde at pH 7. The samples were then washed twice and immersed in 0.1 M phosphate buffer for 1 h. The prepared samples after dehydration with ethanol were coated with gold–platinum alloy before taking their SEM images.

## 3. Results and discussion

### 3.1. Optimization of current density and iron dosages

As the influent wastewater was continuously injected into the EC reactor, the electrical current was uninterruptedly supplied to the reaction tank, ensuring that enough iron is available for the precipitation reactions to take place. To get the optimal amount of current, the amount of iron (mg/L) released with the increase in current density ( $\text{mA}/\text{cm}^2$ ) and its subsequent appearance in the effluent was noted as shown in Fig. 2. The figure shows that the iron liberation was increased with the increase in current density. Meanwhile, the iron contents in the effluent of treated water were also noted. Initially, no iron was detected in the effluent samples when the current density was  $0.2 \text{ mA}/\text{cm}^2$ , revealing that whole metal has been used in the precipitation reactions. But, as the current density increased from  $0.2$  to  $0.4 \text{ mA}/\text{cm}^2$ , the value of total iron in the effluent was observed up to  $0.15 \text{ mg}/\text{L}$ . The current supply was gradually

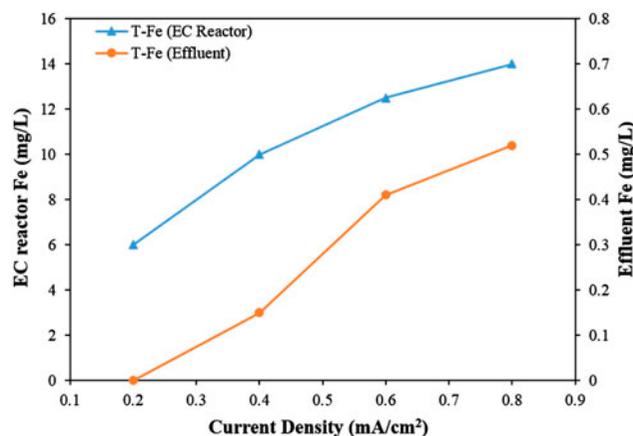


Fig. 2. Fe contents (mg/L) in the EC reactor and effluent samples at different current densities ( $\text{mA}/\text{cm}^2$ ).

increased till the effluent iron attained the values of  $0.52 \text{ mg}/\text{L}$  at  $0.8 \text{ mA}/\text{cm}^2$  and total iron in the reactor was  $14 \text{ mg}/\text{L}$  at that time. Both effluent i.e. Eff-1 and Eff-2 samples showed identical iron values. This point was referred as the optimal point and further studies were conducted on these conditions. During the operation, the samples of effluent for iron were also checked randomly for the smooth running of the plant. The high current density results in the substantial decrease in current efficiency [18], therefore a low current density of  $0.8 \text{ mA}/\text{cm}^2$  was maintained in order to operate for a longer period of time.

### 3.2. Removal of $\text{PO}_4\text{-P}$

The  $\text{PO}_4\text{-P}$  removal efficiencies with the time in modified EC system are shown in Fig. 3. The results

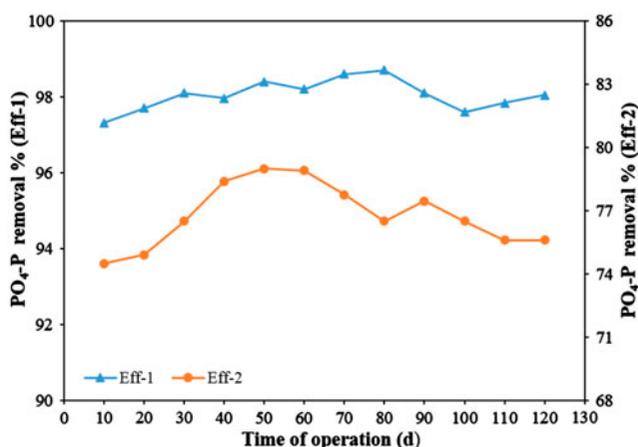


Fig. 3.  $\text{PO}_4\text{-P}$  removal efficiencies (%) of EC coupled with woven membrane (Eff-1) and through settling chamber without membrane (Eff-2) at pilot plant.

showed that the phosphate removal rates by EC coupled with woven membrane were between 97.3 and 98.7% (Eff-1), while in case of Eff-2, it ranged from 74.5 to 78.90%. A very significant difference of nearly 20% in the phosphate removal efficiencies was found by using woven membrane than only performing EC treatment. In our investigations, the pH remained stable at  $7 \pm 0.1$  due to high alkalinity values (120 mg/L) of influent water, which induced enough buffer intensity to the system to stabilize its pH values. The previous studies also displayed that the highest phosphate removal rates by EC were around neutral pH (6.5–7.1) when compared to acidic or basic media [19,20]. The appearance of phosphate in Eff-1 and Eff-2 ranged between 0.1–0.2 and 1.5–20 mg/L, respectively, during the examined period. Stafford et al. [4] mentioned in their results that total phosphorous removal was 30% at current density of  $1 \text{ mA/cm}^2$ , while the maximum removal rate reported by these researchers was 90% at  $4.3 \text{ mA/cm}^2$  current density. On the other hand, in our modified process the removal efficiencies were around 98% at a very low current density of  $0.8 \text{ mA/cm}^2$ . It was very interesting to note that the phosphate removal rates of Eff-2 (74.5–78.90%) were very high as compared with their SS removal efficiencies (52–61%). It revealed that the released iron from the electrode plate preferably attracted the negatively charged phosphate to form iron phosphates. This precipitated material acted as seed particles and agglomerated to large size by attaching other fine SS particles during their settling stage. The high removal also indicated that iron preferably reacted with phosphates instead of other suspended particles, and thus enhanced their removal from the process.

### 3.3. Removal of SS

The difference between the SS removal efficiencies of Eff-1, Eff-2, and  $0.45 \mu\text{m}$  filter samples during the studies is depicted in Fig. 4. The results showed that the SS removal efficiencies were between 97 and 99% for Eff-1 throughout the experimental period. While, it was found between 52 and 61% in Eff-2 samples on the advancement of studies. The high removal efficiency of Eff-1 indicated that the woven fabric membrane worked as an excellent barrier for the contaminants which revealed its high degree of reliability in removing the suspended material from the wastewater. On the other hand, low removal efficiencies of Eff-2 samples were due to the fact that fine particles did not settle down in the settling chamber of EC reactor and remained in the suspension form which eventually passed through the effluent into the discharge stream. Meanwhile, the iron contents in

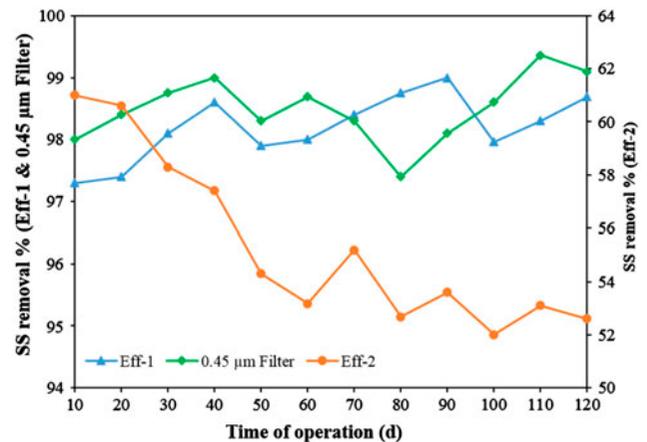


Fig. 4. SS removal efficiencies (%) of Eff-1 (woven fabric membrane), Eff-2 (without membrane), and  $0.45 \mu\text{m}$  filter paper during the operation.

Eff-2 samples were checked and found about  $0.53 \text{ mg/L}$ , when the removal efficiency was 52%. It indicated that all SS could not be removed by employing only EC treatment, even though enough precipitating metal is available in the reactor. The results showed that SS removal efficiencies were almost doubled by using woven membrane than only EC treatment. It can be suggested that the iron mostly removed phosphate, while the majority of SS were removed by woven membrane. The other researchers also highlighted that the release of metals from the electrode preferably reacted with phosphorous and other negatively charged species got deposit around it [14–19]. As, the similar EC conditions were provided for the wastewater treatment, so any difference in the SS removal rates was attributed to the filtration through the woven fiber membrane.

The SS removal efficiency of the woven membrane was also compared by filtering the samples through  $0.45 \mu\text{m}$  filters in the lab. The samples of wastewater from the EC reactor before membrane filtration were regularly collected and filtered through them. This filtrate was analyzed for total SS contents; the results showed that its SS removal rates were almost similar to the woven fabric membrane effluent (Fig. 4) and the filtrate SS contents were less than  $0.35 \text{ mg/L}$ . It is proposed that the woven membrane can be ideally used as a microfiltration membrane in the treatment processes. The results of Eff-1 highlighted that there was no effect on the phosphate and SS removal rates, no matter the flux was increased from 25 to 75 LMH, but a lower trend was noted for Eff-2. The gradual decline of Eff-2 was due to the increase in hydraulic loading rate and precipitates did not have enough time to settle down.

3.4. Performance of woven fabric membrane

The treated water from the reactor was removed by woven membrane at three different flux rates, i.e. 25, 50, and 75 LMH, the total volume processed at these flux rates with time and variation in TMP is shown in Fig. 5. It is evident from Fig. 5 that at 25 and 50 LMH, no increase in TMP took place, so a straight line of TMP touching zero value on the pressure axis can be seen. It may be due to the reason that woven fabric membrane had a three-dimensional open structure and the particles cannot be trapped inside

the membrane pores unlike the generally used hollow fiber membranes. The other reason for uniform running of the membrane was that the contaminant precipitation by electrocoagulant metal increased the overall particle size, which sustained the membrane efficiency which is normally lowered by small particles. But, as the flux was increased from 50 to 75 LMH, a little rise in TMP was noted. The maximum value of TMP at 75 LMH flux was 10 kPa at the close of experiment and the total volume processed at that stage was 226.8 m<sup>3</sup>. The previous studies displayed

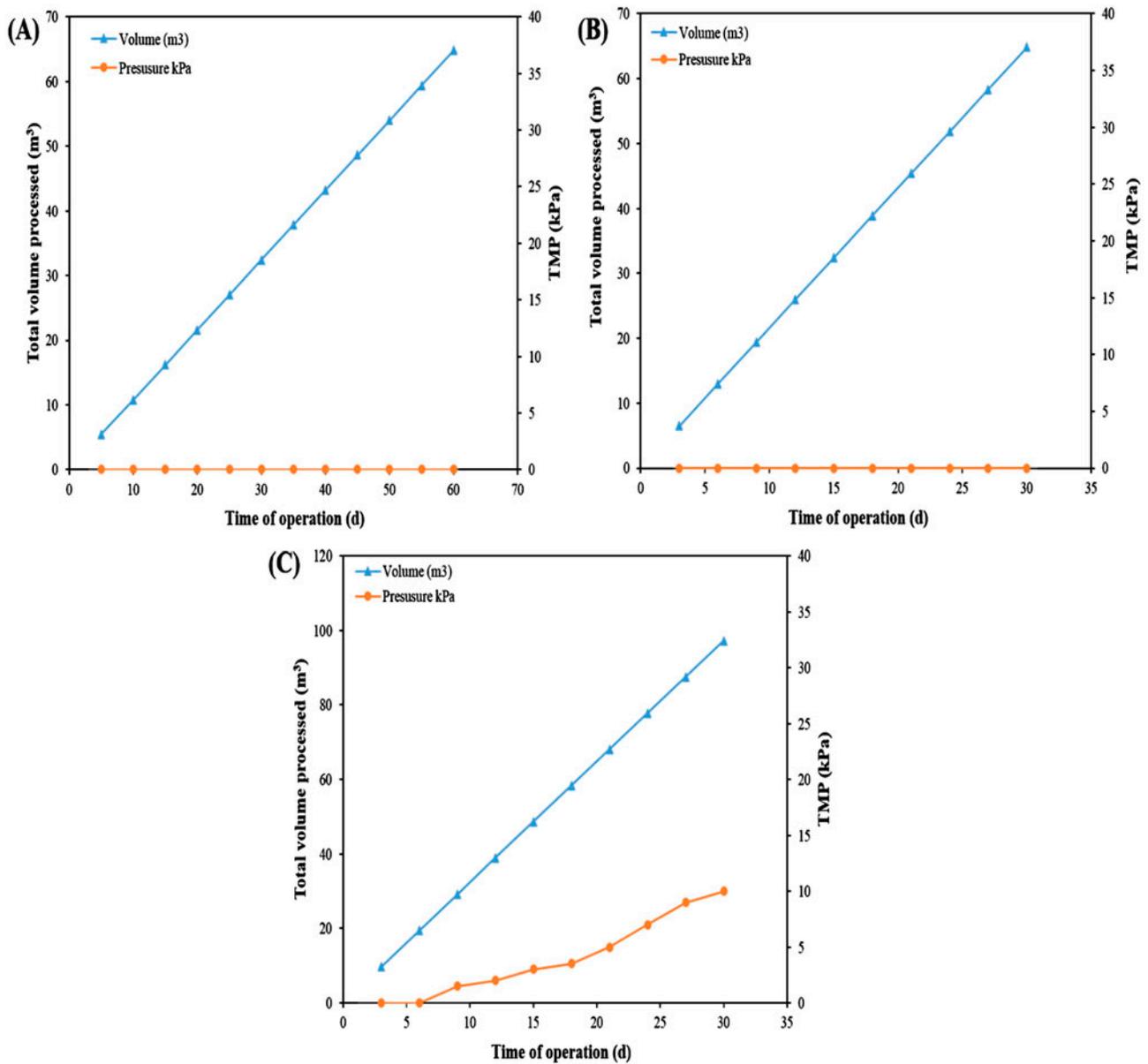


Fig. 5. The total volume processed (m<sup>3</sup>) and the change in TMP (kPa) with time (D) at (A) 25 LMH, (B) 50 LMH, and (C) 75 LMH during the pilot plant studies.

Table 2

Summary of the operating conditions and performance of some Korean full-scale membrane treatment plants

Treatment plant	Membrane material	Surface area (m <sup>2</sup> )	Design flow (LMH)	Operating pressure (atm)	Membrane cleaning method
KMS PF-90	PVDF	90	30–40	3	Physical and chemical
Daewoo eco-star	PVDF	75	40	3	Physical and chemical
DBM-S	PVDF	65	12–20	1	Physical and chemical
Pilot plant (current study)	Woven fabric membrane	1.8	75	1	Physical

that chemical cleaning of microfiltration membranes is only needed if TMP value is increased beyond 40 kPa [12–16,18–21]. Thus, no chemical cleaning was done at any stage of the experiment as the highest TMP value (10 kPa) was still much lower than the reported literature. The aeration provided in the reactor was enough for the physical washing of the woven membrane. The washing by spraying with tap water was only performed when the flux was changed from 25 to 50 and from 50 to 75 LMH. Despite, the changes made in flux rates, the effluent phosphorous and SS results revealed that the effluent quality was stable throughout the experiment as discussed earlier.

The filtration efficiency of woven fabric membrane was compared with some local full-scale membrane treatment plants and the data are given in Table 2. The data provided in Table 2 highlight that the performance of woven membrane was much better than the very costly PVDF membrane used on the full-scale plants. Moreover, the high values of LMH at normal atmospheric pressure and the only need of physical cleaning, make the woven fabric membranes as an excellent alternative for plant scale treatment of the wastewater. Even maintaining a high flux value of 75 LMH than the usual domestic sewage treatment which is performed at 25–30 LMH, no membrane fouling was observed.

The SEM images of physically washed membrane and during operation are shown in Fig. 6. Some precipitated particles can be seen adsorbed on the membrane surface in Fig. 6(A), but still there are many open spaces on the surface which were enough for the passage of effluent through it. These particles were not firmly attached to the surface and when washed with water, all the adhered particles were easily removed and woven membrane was restored in the original shape (Fig. 6(B)).

### 3.5. Significance of the present work

The phosphate removal rate was around 98% at a current density of 0.8 mA/cm<sup>2</sup> in this study as compared to the previously reported literature which showed the maximum removal around 90% at 4.3 mA/cm<sup>2</sup> in the EC system [4]. These results highlighted that the proposed hybrid method was about six times more economical for the contaminant removal. The enhanced removal efficiencies were attributed due to coupling of membrane in the EC reactor. In the routinely employed EC system, the fine particles do not settle down completely in the settling chamber, and passed through the effluent, which lowers the removal efficiencies. Therefore, more current is applied to agglomerate smaller particles, which increases the

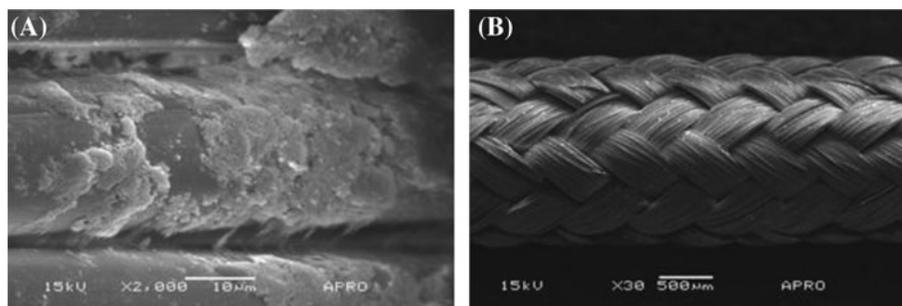


Fig. 6. SEM images of woven fabric membrane (A) during operation at 75 LMH flux and (B) after physical cleaning.

operating cost. However, in case of newly developed method, the fine SS and precipitated particles are retained on the membrane surface and clear effluent is passed through it. The other advantage associated in using this technique than the conventional EC process is that it can be operated in continuous mode as no settling phase is needed due to the insertion of membrane. The cleaning of woven fabric membrane was very easy and only needed flushing with tap water due to its open structure, thus eliminating the use of chemical reagents.

#### 4. Conclusions

From this study, the following conclusions are drawn:

- (1) The phosphate and SS removal efficiencies were about 20 and 62% higher by using the modified process (EC coupled with woven fabric membrane) at a very low current density of 0.8 mA/cm<sup>2</sup> as compared to their removal efficiencies when only EC treatment was employed.
- (2) The effluent quality obtained by filtration through 0.45 μm filter and the woven membrane was identical, which deduced that the woven fabric membrane can be an ideal candidate for microfiltration studies during wastewater treatment.
- (3) A very high flux rate of 75 LMH was achieved without the membrane fouling and the total increase in TMP was only 10 kPa.
- (4) The developed process worked efficiently in the continuous style and also did not involve settling phase, which tremendously decreased the total time required for the water treatment. Moreover, no chemical injection was needed at any stage of the operation for pollutant removal.
- (5) The biggest advantages of using woven membrane for filtration were that it was very cheap, no chemical cleaning needed and showed excellent robustness unlike other commercially available membranes like PVDF etc.

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