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Application of a PAC-Membrane Hybrid System for Removal of Organics from Secondary Sewage Effluent: Experiments and Modelling

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ABSTRACT

As world supplies of clean, fresh water come under increasing pressure and the need for water reuse rises, membrane technology is becoming increasingly important as a possible solution. However, membrane fouling is a major obstacle to the successful operation of the membrane process in wastewater treatment.

In this study, a submerged hollow-fiber membrane with powdered activated carbon (PAC) adsorption was investigated for the removal of organics from secondary sewage effluent from a sewage treatment plant. The use of PAC in the membrane system was found to be very effective,

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not only in removing refractory organics, but also in reducing membrane clogging. A simple mathematical model was developed to predict the effluent quality [in terms of total organic carbon (TOC)] of the submerged membrane–adsorption hybrid system.

Key Words: Adsorption; Hollow fiber membrane; Powdered activated carbon (PAC); Total organic carbon (TOC); Mathematical model; Secondary sewage effluent.

INTRODUCTION

As global supplies of clean, fresh water dwindle and demand for water rises, wastewater reuse is becoming an international focus for the rational use of scarce water resources, and as a means of safeguarding aquatic environments from the harm caused by wastewater disposal. Conventional sewage treatment includes primary treatment to remove the majority of suspended solids, secondary biologic treatment to remove the biodegradable dissolved organics and nitrogen, and tertiary treatment to remove most of the remaining organics, solids, and pathogenic microorganisms. Although the effluent from this treatment can be discharged into a waterway, it cannot, for reasons of health and safety, be recycled. To obtain water of recyclable quality, physicochemical processes such as flocculation, sedimentation, filtration, and adsorption were initially tried. With technologic advances and the ever-increasing stringency of water quality criteria, membrane processes have become a more attractive solution to the challenge of water reuse. Although membrane processes, such as reverse osmosis and nanofiltration could in theory remove all pollutants, including dissolved organics, their operational costs are high because of high-energy requirements and membrane fouling. Micro- or ultrafiltration are cost-effective options, but they cannot remove dissolved organic matter due to their relatively larger pore sizes. In a number of water reuse applications, mainly in the late 1990s, ultrafiltration or microfiltration were combined with biologic processes. For example, in a water-mining project in Canberra, Australia, biologic filtration was combined with continuous microfiltration. The Lyonnaise des Eaux built a number of water recycling plants in France (for both sewage and industrial wastes) that use an activated sludge process followed by ultrafiltration. Most of these plants use membranes in the external loop, and this has a high-energy demand. To reduce the energy requirement, membrane clogging, and to simplify the process, a bioreactor with submerged membranes is used. A series of semiproto scale plants have been designed by industries and academic institutions in Australia, Japan, France, and the United States. These have been successfully tested on a long-term basis.[1] Submerged membrane units are simple to operate and suffer minimum membrane clogging.

However, biologic degradation may be disrupted in submerged membrane bioreactors if wastewater is contaminated with toxic and other matter, which is not readily biodegradable. Submerged membrane bioreactors carry out the entire treatment activity (namely biodegradation, solid–liquid separation, and sludge accumulation and withdrawal) in a single unit. This can pose operational problems. To facilitate the removal of refractory organics, the submerged membrane reactor may be dosed with adsorbents. This process is physicochemical in nature and can be easily controlled. Owing to the need to remove refractory organics cheaply and to reduce membrane clogging (fouling), the addition of powdered activated carbon (PAC) to micro/ultra filtration systems (known as an adsorption–membrane filtration hybrid system) is emerging as a highly promising water and wastewater treatment technology.[2–8] In this process, the pollutants (particularly the dissolved organics) are first adsorbed onto PAC, so greatly reducing the direct loading of dissolved organic pollutants onto the membrane. The use of PAC in the system not only increases the permeate flux (effluent filtration rate) through the membrane, it also prolongs its life by minimizing fouling by organics.

The initial decrease in the permeate flux in microfiltration is mainly due to rapid, irreversible adsorption of organic substances onto the membrane surface.[9] Therefore, the preadsorption of organics prior to passage of the wastewater solution through the membrane promises to be very effective in reducing membrane fouling.

Most previous studies of the PAC-membrane hybrid system have concentrated on the removal of a targeted pollutant. Kim et al.[6] found the PAC-membrane hybrid system very effective in removing coliphase-Qβ and total organic carbon (TOC) from the synthetic secondary wastewater. They used a submerged hollow fiber membrane with a predetermined PAC dose, and found 99.9% and 95% removal of coliphase-Qβ and TOC, respectively, for a PAC dose of 40 g/L. The system operated successfully without significant fouling for a long period. Seo et al.[3] found that the biologic powdered activated carbon (BPAC)-MF hybrid system could remove 83% of TOC and 99.99% of coliphase-Qβ with a PAC dose of 20 g/L. They allowed a contact time of 1 h for PAC with the wastewater.

It is vital that the effectiveness of the hybrid system be evaluated using biologically treated sewage effect before it can be applied for water reuse. Further, the initial adsorption of organics by this system has to be optimized through the correct choice of PAC type and its dose. In this study, wood-based PAC was used as an adsorbent in the submerged membrane system to evaluate the organic removal efficiency of the hybrid system. Secondary sewage effluent (from the secondary sedimentation tank) from a sewage treatment plant in Sydney was used as the wastewater. The main objective of the study is to evaluate the effectiveness of the PAC-membrane hybrid system for organic removal in short-term and long-term basis, and to predict the performance of the hybrid system by a simple model. In this study, a simple mathematical model for the submerged hollow fiber membrane hybrid system was developed to predict effluent quality from the system. The effects of adsorption and membrane parameters in the mathematical model were also analyzed.

CONCEPTUAL MODEL FOR MEMBRANE HYBRID SYSTEM

The application of the PAC-membrane hybrid system in water and wastewater treatment processes is relatively new. In the PAC-membrane hybrid system, dissolved organic compounds are adsorbed onto PAC particles, and the PAC, together with its adsorbed organics, are eventually separated by the membrane filtration process. Previous studies have shown that the addition of PAC to the membrane filtration system is a simple and cost-effective way to remove dissolved organic (natural and synthetic) compounds.[10,11] The treatment efficiency of the hybrid system depends on reactor configuration, operating modes, carbon dose and adsorptive capacity, and influent characteristics. Depending on the operational mode, a continuous-flow stirred tank reactor (CSTR) and a plug-flow reactor (PFR) are the two main models used in the design of a membrane hybrid system.

Extensive research has been done into the mathematical modeling of the adsorption kinetics of dissolved organic compounds by PAC.[12–14] Najm[15] applied a homogenous surface diffusion model (HSDM) and Freundlich isotherm to model the CSTR and PFR modes of the adsorption system. It is important to use the adsorption kinetics of the PAC in the PAC-membrane hybrid system. However, there are very few published studies on the integration of PAC adsorption modeling into a membrane filtration system. Campos et al.[16,17] utilized the HSDM model with no external mass transfer limitations and developed an integrated mathematical model to predict the removal of organic compounds in the PAC-membrane hybrid system. They considered four cases of adsorption: (1) a membrane reactor fed with step inputs of fresh PAC, (2) a CSTR-membrane system fed with step inputs of fresh PAC, (3) a PFR-membrane system fed with step inputs of fresh PAC, and (4) the membrane reactor fed with pulse inputs of fresh PAC. Model predictions were based on a single set of equilibrium and kinetic parameters independently determined from the results obtained in bench-scale experiments.

Kim et al.[6] used the PAC-MF (submerged membrane) system to evaluate the removal efficiency of coliphase Qβ and organic matter from a synthetic secondary effluent. They assumed a first-order driving force model to predict the Qβ removal efficiency of the PAC-MF system.

These models are capable of predicting the system's organic removal efficiency. However, they do not incorporate the characteristics of the influent or the membrane properties, such as different organic and hydraulic loadings, and packing density of the membrane. These parameters are very likely to change during the operation of a membrane treatment system.

In this study, a simple mathematical model using the concept of a continuous flow stirred tank reactor (CSTR) was developed. Membrane packing density (A_M/V_M) and membrane correlation coefficient (MCC) were incorporated into adsorption model. Although, the MCC is just and empirical and fitting parameter, it gives a new dimension in modeling the long-term operation of the hybrid system. The model can be further expanded to incorporate the biodegradation of the organics by the microbial community on the adsorbent. The latter is likely to be the most influential mechanism in the long-term successful operation of the hybrid system. Since the filtration flux is very low, the decline in flux is not considered in this model.

The mass balance of the PAC-MF system can be described by eq. (1)

$$\frac{dC_b}{dt} = \frac{Q}{V}\cdot(C_0 - C_b) - \frac{M}{V}\cdot\frac{dq}{dt} - \frac{A_M}{V_M}\cdot MCC\cdot C_b - BIO \tag{1}$$

where, C_b is the organic concentration in the bulk phase in the reactor (mg/L), Q the flow rate (m^3/s), V the volume of the bulk solution in the reactor (m^3), C_0 the organic concentration in the feed tank (mg/L), M the weight of PAC used (g), A_M the surface area of the membrane (m^2), V_M the volume of the membrane (m^3), MCC the membrane correlation coefficient, BIO the biodegradation by the microbial community.

The second term, [(M/V)·(dq/dt)], on the right hand side of eq. (1) represents the adsorption of the organics onto PAC in suspension, and the third term, [(A_M/V_M)·MCC·C_b], the adsorption onto the PAC layer deposited on membrane surface. Here the term, A_M/V_M represents the packing density of the membrane. The membrane correlation coefficient (MCC) is an empirical coefficient that incorporates both adsorption of organics on membrane surface and retention of PAC (associated with organics adsorbed on it) on membrane. The higher the value of MCC, the more efficient the organic removal by the membrane hybrid system. The fourth term [BIO] of the equation is introduced to account for the removal of organics by microbial biodegradation. Since only short-term experiments were carried out in this study, the term BIO is omitted from the model. Further research is required to extend the model by incorporating the biodegradation of organics in the system.

The mass transfer rate inside the PAC was described by a linear driving force approximation (LDFA) model, as shown in eq. (2). The model assumes that the rate of mass transfer is directly proportional to the concentration gradient developed between the surface concentration on the PAC and the average adsorbed phase concentration of the adsorbate.[18]

$$\frac{dq}{dt} = k_s \cdot (q_s - q) \tag{2}$$

where, q is the adsorbed phase organic concentration at any time (t), q_s the equilibrium adsorbed phase organic concentration, and k_s the solid mass transfer coefficient.

The initial conditions for mass balance in bulk phase and solid phase are:

$$C = C_0$$

and

$$q = 0$$

respectively.

The adsorption condition equilibrium is represented by Freundlich isotherm, eq. (3).

$$q_s = K_F \cdot C^{\frac{1}{n}} \tag{3}$$

Here, K_F and n are the Freundlich adsorption coefficient and exponential coefficient respectively, and C is the equilibrium bulk phase organic concentration.

The differential equations were solved using the subroutine, DVODE.[19] The sensitivity of the model for different adsorption and membrane parameters, and the experimental results with model prediction, are discussed in the following section.

Sensitivity Analysis of the Adsorption Dynamics Model

The model developed for the membrane hybrid system was tested for various parameters such as membrane correlation coefficient, solid mass transfer coefficient, filtration flux, and influent organic concentration used in the model (Figs. 1 to 4). The performance of the model was found to depend mainly on membrane correlation coefficient (MCC) and the filtration flux. The higher the value of MCC, the better the organic removal efficiency of

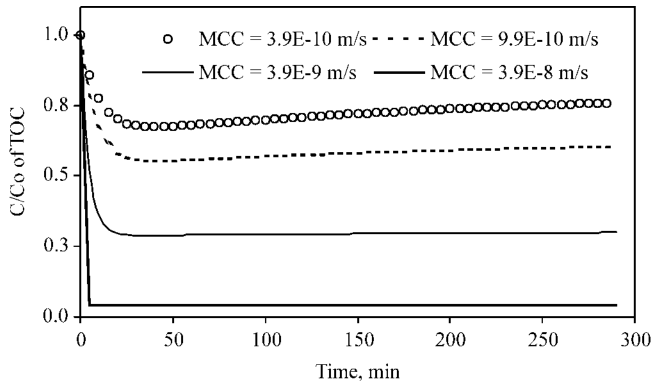

Figure 1. Effect of membrane correlation coefficient on the effluent TOC. PAC dose = 200 mg/L; influent TOC = 2.6 mg/L; $k_s = 1.6E-6\,1/s$; filtration flux = 3×10^{-3} L/s/m^2; $K_F = 13.2$; $1/n = 1.5$; C = effluent TOC concentration; mg/L and C_0 = influent TOC concentration, mg/L.

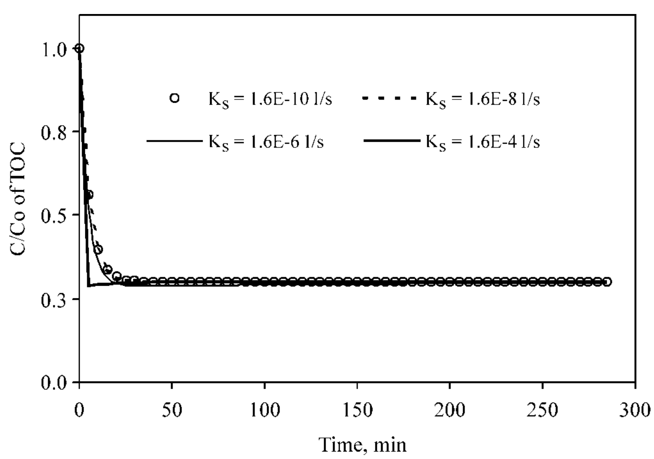

Figure 2. Effect of solid mass transfer rate (k_s) on the effluent TOC. PAC dose = 200 mg/L; influent TOC = 2.6 mg/L; MCC = $3.9E-9$ m/s; filtration flux = 3×10^{-3} L/s/m^2; $K_F = 13.2$; $1/n = 1.5$; C = effluent TOC concentration; mg/L and C_0 = influent TOC concentration.

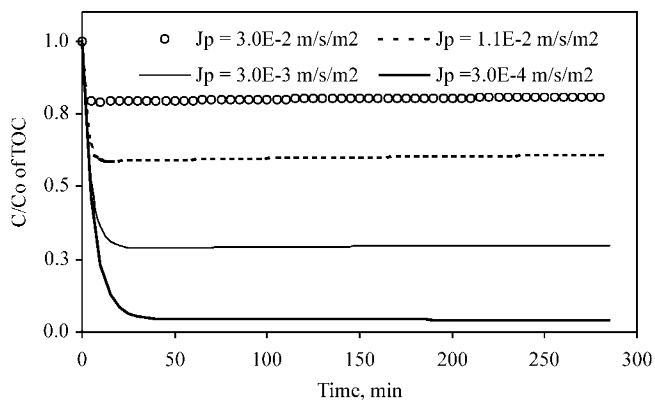

Figure 3. Effect of filtration flux on the effluent TOC. PAC dose = 200 mg/L; influent TOC = 2.6 mg/L; MCC = 3.8×10^{-9} m/s; $k_s = 1.6 \times 10^{-6}$ l/s; $K_F = 13.2$; 1/n = 1.5; C = effluent TOC concentration; mg/L and C_0 = influent TOC concentration.

the system. However, as anticipated, the organic removal efficiency of the system decreases when the filtration rate is increased. The model was sensitive neither to the solid mass transfer coefficient (k_s) nor to the influent organic concentration (C_0) for the wide range of values tested (see Figs. 2 and 4).

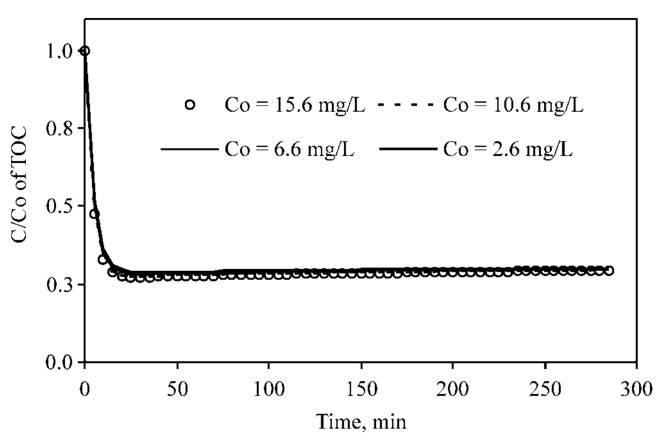

Figure 4. Effect of influent TOC concentration on the effluent TOC. PAC dose = 200 mg/L; filtration flux = 3×10^{-3} L/s/m^2; MCC = 3.9×10^{-9} m/s; $k_s = 1.6 \times 10^{-6}$ l/s; $K_F = 13.2$; 1/n = 1.5; C = effluent TOC concentration; mg/L and C_0 = influent TOC concentration.

Previous studies by the authors showed that the isotherm parameters and the value of k_s are directly related to the initial organic concentration (C_0) and the adsorbent dose respectively.[18] In the sensitivity analysis, both the PAC dose and the influent organic concentration were kept constant. Since, in this analysis, the PAC dose and the isotherm parameters are constant, no direct effect of C_0 and k_s could be observed.

EXPERIMENTAL INVESTIGATION

Three types of experimental studies were carried out: (1) adsorption equilibrium, (2) batch kinetics, and (3) PAC-membrane hybrid microfiltration with submerged hollow fiber membrane. The adsorption equilibrium and the batch kinetics studies were carried out to determine the adsorption characteristics of PAC in the membrane hybrid system.

Adsorption Equilibrium

Adsorption equilibrium experiments were conducted using 250 mL of secondary effluent solution from a sewage treatment plant in flasks. The amount of PAC used for the study varied from 0.1 g to 1.7 g. The flasks were shaken continuously for 7 days at 130 rpm at 25°C (lab conditions). Total organic carbon (TOC) was measured using the UV-persulphate TOC analyzer (Dohrmann, Phoenix 8000). The PAC used in the experiments was washed three times with distilled water and dried in the oven at 103.5°C for 24 hours. It was kept in a desiccator before using in the experiments.

The TOC concentration of the secondary effluent (from a sewage treatment plant in Sydney) used in this study remained fairly stable between 3 and 4.5 mg/L. The physical properties of the PAC are shown in Table 1.

The adsorption equilibrium result of the secondary effluent from a sewage treatment plant is shown in Fig. 5. The Freundlich isotherm was used to describe the isothermal adsorption behavior of the system.

Adsorption Batch Kinetics Experiments

The batch experiments were conducted using 2 L of the wastewater solution at the laboratory. The PAC dose was varied from 100 to 400 mg/L and the stirring speed was maintained at 100 rpm.

Table 1. Physical properties of the PAC used in the study.

Specification	Estimated value
Iodine number, mg/g min	900
Maximum ash content	6% max
Maximum moisture content	5% max
Bulk density, kg/m^3	290–390
BET surface area, m^2/g	900
Nominal size, m	80% min finer than 75 μm
Average pore diameter, Å	25.3

Wood-based PAC was used to study the mass transfer rate from the wastewater bulk solution to the PAC surface. The mass transfer rate was calculated by fitting the experimental data using the linear driving force approximation (LDFA) model. The solid mass transfer coefficient (k_s) in the secondary sewage solution was estimated to be almost constant at 12.5×10^{-5} l/s for the PAC dose of 100 to 400 mg/L. The TOC removal efficiency of the PAC did not increase in proportion when the PAC dose was increased from 100 to 400 mg/L. The experimental results and the LDFA model prediction are shown in Fig. 6.

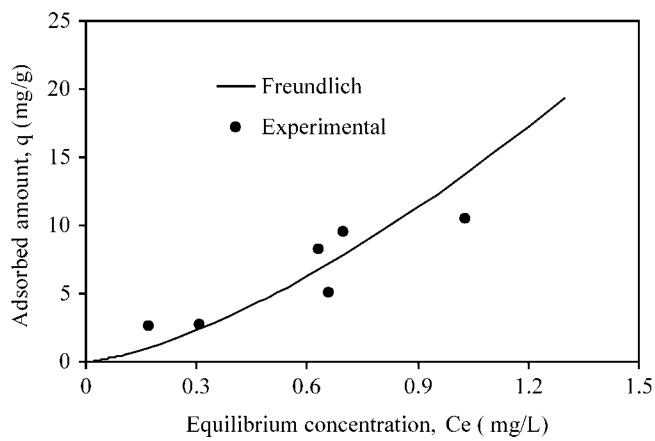

Figure 5. Overall adsorption isotherm of secondary effluent from St. Marys sewage treatment plant. Initial TOC = 3.5 mg/L; K_F = 13.2; 1/n = 1.5.

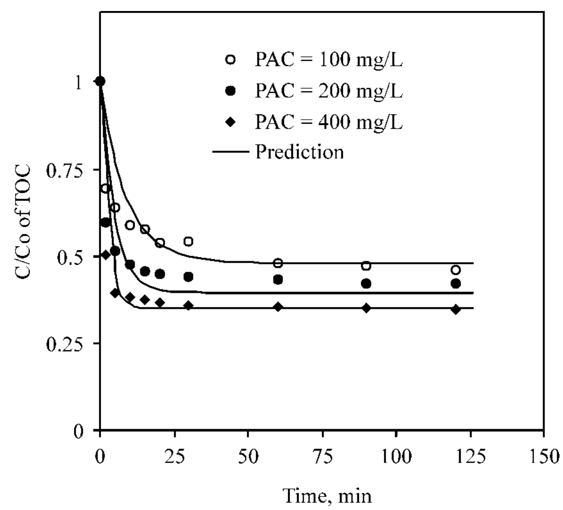

Figure 6. Mass transfer at different PAC dose. Stirring speed = 100 rpm; average initial TOC concentration of the secondary sewage = 3.6 mg/L; C = effluent TOC concentration; mg/L and C_0 = influent TOC concentration; $k_s = 12.5 \times 10^{-5}$ l/s.

Submerged PAC-Membrane Adsorption Hybrid System

The schematic diagram of the submerged hollow fiber microfiltration system is shown in Fig. 7. Wastewater was pumped into the reactor (9 L) using a feed pump to control the influent and effluent flow rate. A predetermined amount of PAC was added into the tank (pulse input) to adsorb the dissolved organic substances, which was subsequently separated by the membrane filtration imposed by the suction pump. The inflow and outflow of the wastewater solution were kept equal to maintain a constant volume of wastewater in the reactor. A pressure gauge was used to measure the transmembrane pressure of the hybrid system. An air diffuser was used to maintain the PAC in suspension. This also provided the dissolved oxygen necessary to maintain the microbial community in the reactor when the system is used for long-term operation. However, this study was limited to short-term adsorption-membrane filtration only. The loss of volatile organic compounds (VOCs) due to aeration was neglected as the wastewater used in this study was biologically treated and hence well stabilized. Table 2 shows the physical and chemical properties of the hollow fiber membrane used in this study.

The effects of PAC dose on the TOC concentration profile of the effluent of the hybrid system are shown in Figs. 8 and 9. As expected, the organic

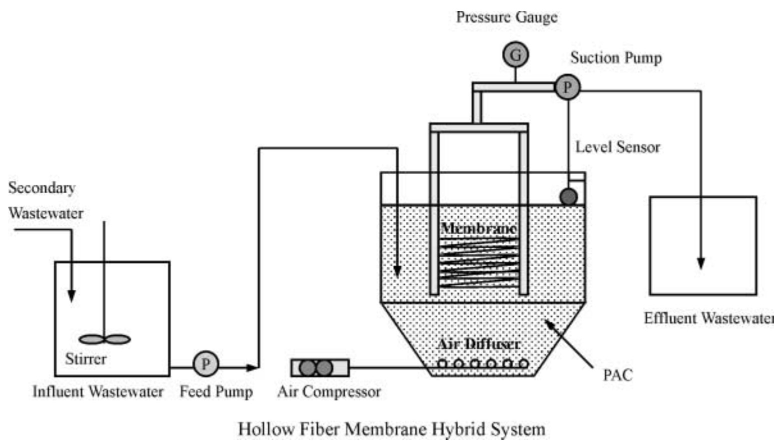

Figure 7. Schematic diagram of submerged hollow fiber membrane filtration hybrid system.

removal efficiency of the system increased with the increase in PAC dose. The solid mass transfer coefficient of the system was observed to be insensitive to the PAC dose and remained almost constant. However, the membrane correlation coefficient of the system was increased when higher doses of PAC were fed into the system. The relationship between PAC doses, filtration flux, and membrane correlation coefficient (MCC) is shown in Fig. 10. The MCC was also found to increase marginally with the increase in filtration flux.

Continuous operation of the PAC-membrane hybrid system for 47 days showed that the system can be operated effectively for a long time with consistent organic removal efficiency and little or no membrane clogging (Fig. 11). When higher doses of PAC were fed into the reactor (in pulse input mode), the hybrid system could achieve higher removal and longer periods of

Table 2. Physical and chemical properties of the membrane.

Properties	Hollow fiber membrane
Total surface area (m^2) (320 fibers with 12 cm length)	0.05
Pore size (μm)	0.1
Material	Polyethylene
Inner diameter (mm)	0.27
Outer diameter (mm)	0.41

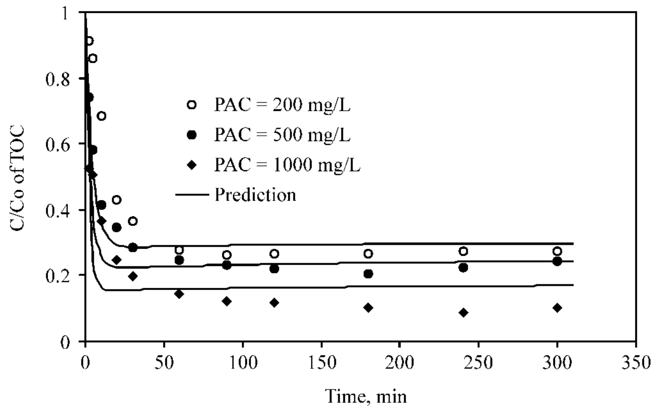

Figure 8. Effect of PAC dose on the performance of the submerged membrane at filtration flux of 3×10^{-3} L/s/m^2. Average influent TOC of secondary sewage = 2.9 mg/L; C = effluent TOC concentration; mg/L and C_0 = influent TOC concentration.

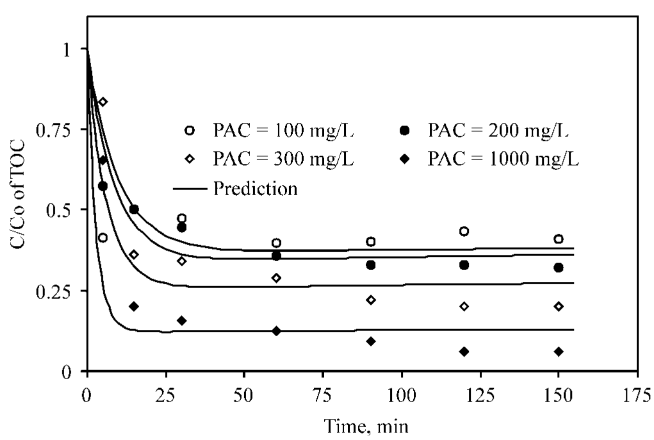

Figure 9. Effect of PAC dose on the performance of the submerged membrane at filtration flux of 1.7×10^{-3} L/s/m^2. Average influent TOC of secondary sewage = 3.0 mg/L; C = effluent TOC concentration; mg/L and C_0 = influent TOC concentration.

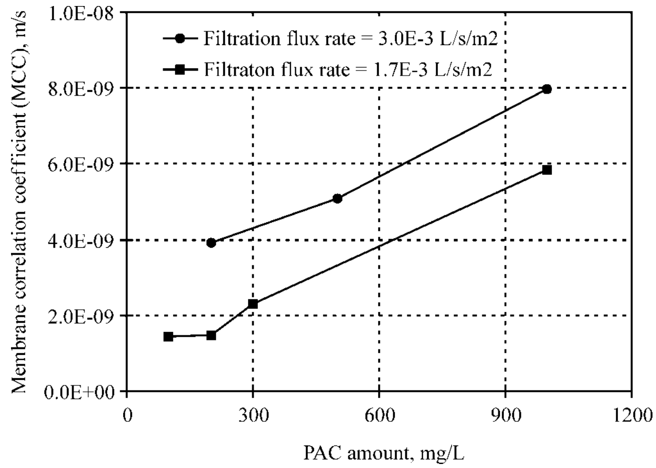

Figure 10. Relationship between membrane correlation coefficient (MCC), filtration flux rate, and PAC dose. Average influent TOC of secondary sewage = 2.95 mg/L.

operation. Kim et al.[6] found that the organic removal efficiency of their submerged hybrid system from a synthetic secondary effluent was more than 95% and 90% for the PAC doses of 40 g and 10 g, respectively, for more than 40 days. Here, biologic degradation was also significant as an organic removal of 73% was observed without adding PAC. In addition to the improved water

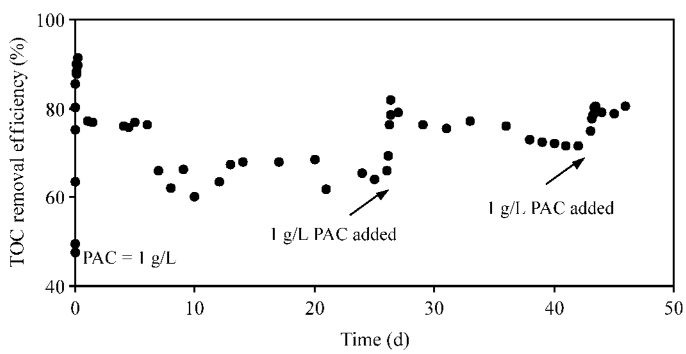

Figure 11. Long-term performance of the submerged membrane hybrid system. Average influent TOC of the secondary sewage = 3.0 mg/L; C = effluent TOC concentration; mg/L and C_0 = influent TOC concentration.

quality, it was also observed that high PAC dosages in the membrane reactor substantially reduced and slowed membrane fouling.

CONCLUSION

The submerged PAC-membrane hybrid system was found to be highly effective in removing dissolved organic substances from secondary effluent of a sewage treatment plant. The system has tremendous potential for the long-term application in the treatment and reuse of wastewater. During the initial stages of the operation, organic removal is mainly due to adsorption onto PAC, but after a few days of operation, when the adsorption capacity of the PAC is reached, the degradation of organics appears to be due to biologic degradation caused by the microbial communities, which grow in the suspension of the reactor, on the PAC, and on the membrane surface. There is also biodegradation of organics adsorbed onto the PAC and a subsequent adsorption on the renewed PAC.

A semiempirical mathematical model was developed for the PAC-submerged hollow fiber membrane hybrid system, which takes into account the adsorption of organics onto the PAC and onto the membrane surface, and separation of PAC (with organics adsorbed on it) by the membrane. A new term, membrane correlation coefficient (MCC) was introduced to describe both the adsorption of organics onto the membrane surface and retention of PAC on membrane. The MCC and the filtration flux were found to be the main model parameters controlling the quality of effluent from the system. The greater the value of MCC, the better the system's organic removal efficiency. The MCC value, on the other hand, was found to increase in proportion to the increase in the PAC dose to the system. The model could predict the performance of the hybrid system successfully for the range of PAC dose of 100 mg/L to 1000 mg/L. Since only the short-term experiments were conducted in this study, the biologic degradation of organics has not been included in the model. This will be necessary to predict the long-term efficiency of the system.

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