Treatment of Formaldehyde-Containing Wastewater Using Membrane Bioreactor

Chalor Jarusutthirak[1]; Kamolchanok Sangsawang[2]; Supatpong Mattaraj[3]; and Ratana Jiraratananon[4]

Abstract: Performance of a membrane bioreactor (MBR) in removal of formaldehyde from synthetic wastewater was investigated. Batch tests for biodegradation of formaldehyde indicated that bioreactors containing acclimated sludge were able to remove up to 99.9% of the formaldehyde from solution. The 12-L MBR was equipped with a submerged hollow-fiber ultrafiltration (UF) membrane with 0.85 m^2 filtration area. The unit was operated at a hydraulic retention time of 10 h in aerobic mode with formaldehyde as the sole carbon source for microbial growth. The results revealed that the MBR reduced formaldehyde concentration from 526 ± 30 to a 1.39 ± 0.73 mg/L, corresponding to a removal efficiency of $99.73 \pm 0.14\%$. Increasing solid retention time (SRT) resulted in an increase in mixed liquor suspended solids (MLSS), leading to improved efficiency in removal of formaldehyde from the MBR. Flux decline during MBR operation was caused by accumulation of MLSS on the membrane surface. SRT did not affect flux decline, but did affect flux recovery after cleaning. Long SRT (60 days) led to greater flux recovery than shorter SRTs (30 and 10 days). **DOI: 10.1061/(ASCE)EE.1943-7870.0000430.** *© 2012 American Society of Civil Engineers.*

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Introduction

Formaldehyde is commonly used in industrial processes for a variety of products, for example, urea–formaldehyde adhesives, aminoplastic resins, textile, dyes, paper, leather, and glass mirrors (Glancer-Soljan et al. 2001; Campos et al. 2003). It is frequently found in wastewaters and waste gases causing environmental pollution. Formaldehyde-containing wastewater has been found to be toxic to aquatic ecosystems because it acts as a substrate inhibitor in several biochemical functions and biotransformation (Moteleba et al. 2002; Eiroa et al. 2005). Therefore, elimination of formaldehyde prior to discharge into the natural receiving water is necessary to preserve environmental quality.

Formaldehyde-containing wastewater can be treated by physicochemical processes, for example, ozonation (Garrido et al. 2000), hydrogen peroxide (Kajitvichyanukul et al. 2006), and catalytic wet oxidation (Silva et al. 2003); and biological processes, for example, anaerobic systems and activated sludge processes (Garrido et al. 2001; Lotfy and Rashed, 2002; El-Sayed et al. 2006). Formaldehyde removal by biological methods is generally preferred because of the lower costs and the possibility of complete mineralization. Formaldehyde has been observed to be readily biodegradable in both aerobic and anaerobic systems at low concentrations. However, above a threshold value, it becomes highly toxic (Garrido et al. 2001). Not many microorganisms can degrade formaldehyde; this is attributed to its toxic effect on parts of bacterial cells (spores, cell walls, and compounds with amino groups). Formaldehyde concentrations greater than 250 mg/L inhibit microbial activity in aerobic wastewater treatment plants. Some strains of microorganisms have been reported to be capable of degrading formaldehyde. *Pseudomonas* species degrade formaldehyde through formaldehyde dismutase, whereas the yeasts *Hansenula* species and *Candida* species use formaldehyde dehydrogenase and formate dehydrogenase to degrade formaldehyde. A basic intermediate in formaldehyde biodegradation is formic acid, which is known to be readily biodegradable and can be further degraded by general microorganisms (Glancer-Soljan et al. 2001; El-Sayed et al. 2006).

Although previous studies demonstrated the feasibility of treating wastewater containing formaldehyde, it was observed that relatively low conversion rates, bacteria washout, and system instability occurred in conventional activated sludge processes, leading to high concentration of formaldehyde and other organic compounds in the effluent (Eiroa et al. 2005; El-Sayed et al. 2006). To enhance the stability and performance of the activated sludge process in the removal of formaldehyde, acclimated bacteria need to be retained in the system. The membrane bioreactor (MBR) system was introduced to maintain better control of the biosolids that are usually washed out of biological systems with secondary clarifiers (Artiga et al. 2005; Melin et al. 2006). The MBR system consists of an activated sludge bioreactor coupled with a submerged ultrafiltration (UF) membrane. The UF membrane not only has the advantage of retaining formaldehyde-acclimated biosolids in the bioreactor, but also replaces the conventional sedimentation

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process. As a result, the MBR system has proven to be a promising wastewater treatment alternative in terms of effluent quality, low/zero sludge production, high loading rate capacity, reliability, and compactness compared with conventional activated sludge processes (Wen et al. 2004; Galil and Levinsky, 2007; Qin et al. 2007).

The aforementioned studies reported the advantage of the MBR in containment of acclimated sludge for formaldehyde removal. There has been no report on removal of formaldehyde using the MBR. Hence, this work was aimed at employing the MBR system in the treatment of formaldehyde-containing wastewater. The ability of acclimated sludge to remove formaldehyde was investigated in batch tests. The effects of formaldehyde concentration, pH, and biosolid content [in terms of mixed liquor suspended solids (MLSS)] were studied. Then, performance of the MBR in removal of formaldehyde from synthetic wastewater was evaluated. Effects of solid retention time (SRT) on formaldehyde removal efficiency and membrane flux decline were also studied. The results from this study can be applied to the treatment of industrial wastewater containing formaldehyde to yield high-quality effluent for sustainable reclamation and reuse of industrial wastewater.

Materials and Methods

Source of Wastewater

Formaldehyde was the sole carbon source for the biological system. Synthetic wastewater was prepared from 38% (w/v) formaldehyde stock solution. The concentration of formaldehyde introduced into MBR system was made approximately 500 mg/L by diluting with deionized (DI) water. Nutrients required for microbial growth and metabolism were added to the synthetic wastewater. The COD:N:P ratio was maintained at 200:5:1 (Eiroa et al. 2004). In this research, ammonium sulfate ($(NH_4)_2SO_4$) was used as the nitrogen source, whereas potassium dihydrogen phosphate (KH_2PO_4) and disodium hydrogen phosphate (Na_2HPO_4) were used as phosphate sources and a buffer system at pH 7.

Experimental Setup

Batch Tests

Batch tests were conducted in laboratory-scale reactors made of 2.5-L amber glass bottles with an effective volume of 2 L. Feed air was continuously supplied by an air pump through air diffusers. Seed sludge, obtained from the wastewater treatment plant of a urea–formaldehyde resin manufacturer, was acclimated in synthetic wastewater before the experiments.

All batch experiments were performed at room temperature (~30°C). The pH in the reactor was maintained at 7.0–7.5 except when the effect of pH was being tested. Dissolved oxygen was maintained above 2 mg/L at all times. Effects of formaldehyde concentration, pH, MLSS, and contact time on formaldehyde removal efficiency were investigated.

Membrane Bioreactor

The submerged MBR was acrylic and had an effective volume of 12 L (Fig. 1). The membrane module was installed vertically in a screened column. It consisted of polysulfone hollow-fiber membranes with a molecular weight cutoff in the UF range and a filtration area of 0.85 m^2. Air bubbles were continuously supplied through air diffusers. One diffuser was located underneath the membrane module to scour the membrane surface; the other two diffusers were located at the bottom of the tank to supply oxygen for biomass growth and provide turbulence in the reactor. Synthetic wastewater, as an influent, was continuously fed into the reactor at the rate of 1.2 L/h, corresponding to a hydraulic retention time (HRT) of 10 h.

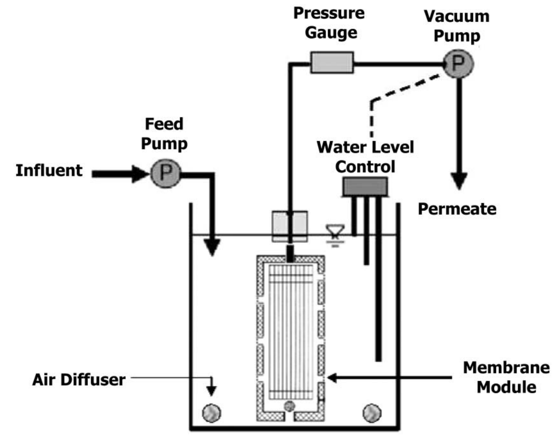

Fig. 1. Schematic diagram of the membrane bioreactor

Seed activated sludge, obtained as described earlier, was used to inoculate the system. During the acclimation period, the formaldehyde concentration in the influent was increased stepwise 100 to 500 mg/L and then maintained at approximately 500 mg/L. Each organic loading rate was maintained in the system for a period at least three times greater than the applied HRT to reach steady-state conditions. Permeate was obtained using a vacuum pump, which was controlled by a water level sensor to maintain a constant water level in the bioreactor.

The effects of SRT on MBR performance in formaldehyde removal and membrane fouling were investigated. SRT was varied from 10, to 30, to 60 days by daily wasting mixed liquor sludge from the reactor. The volumes of mixed liquor wasted were estimated as described by Grady et al. (1999). The waste stream volumes needed to maintain SRTs of 10, 30, and 60 days were 1.2, 0.4, and 0.2 L/day, respectively.

Permeate flux, representing permeate volume per membrane area per filter time, was determined to assess membrane fouling. As suction pressure was not maintained constant, specific flux was reported as a function of permeate flux per suction pressure, in units of L/m^2-h-kPa (LMH/kPa).

Water samples were taken at different points, including the influent tank, aeration tank, and permeate tank. All samples, except UF permeates, were filtered through 0.45-μm filter paper, prior to analyses. Formaldehyde, total organic carbon (TOC), MLSS, dissolved oxygen (DO), and pH were measured every other day on a regular basis. All analyses were carried out in duplicate. Statistical analysis using ANOVA was performed to analyze experimental results. Percentage removal was expressed as means ($\pm$ standard deviations). After 5–7 days of operation, the membrane module was removed for hydrodynamic cleaning followed by caustic (0.001 M NaOH) and acid (0.001 M citric acid) cleaning. Before it was replaced in the MBR system, the specific flux of pure water for the cleaned membrane was measured to determine the initial flux of the membrane.

Analytical Methods

Formaldehyde was determined according to the Hantzsch reaction (Nash 1953). A properly diluted formaldehyde solution was supplemented with acetyl acetone and ammonium acetate to form a colored compound. This compound was analyzed at 412 nm with a UV/Vis spectrophotometer (Thermo Electron, England). The minimum detection limit of this method was less than 1 mg/L (Helrich 1990). TOC was analyzed with a combustion method

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using a TOC analyzer (Shimadzu, Japan). MLSS, DO, and pH were evaluated according to standard methods [American Public Health Association (APHA)/American Water Works Association (AWWA)/Water Environment Federation (WEF)].

Results and Discussion

Effects of Formaldehyde Concentration and Contact Time on Formaldehyde Removal

To study the potential of formaldehyde biodegradation, formaldehyde concentrations in the range 100–1,000 mg/L were introduced into bioreactors in batch tests. MLSS concentration was approximately 500 mg/L in each reactor. The pH was maintained at 7 using phosphate buffer. Samples were taken at various contact times.

Fig. 2 shows that formaldehyde concentration in the solution decreased gradually as contact time increased. At a contact time of 48 h, formaldehyde was treated until its concentration decreased below 1 mg/L, which is a local industrial effluent standard. The biological process with acclimated microorganisms was able to decrease formaldehyde concentration from 1,000 mg/L to less than 1 mg/L, that is, greater than 99.9% removal, at a contact time of 48 h. This finding proved that formaldehyde was biodegraded by microorganisms in acclimated sludge. Similar results were reported by Glancer-Soljan et al. (2001), who explained that some acclimated strains of bacteria were able to survive in the formaldehyde-contaminated environment, for example, *Pseudomonas putida*, *Pseudomonas cepacia*, *Hansenula* species, and *Candida* species. As a carbon substrate for their growth, these strains use dehydrogenase, an enzyme that can degrade formaldehyde (Glancer-Soljan et al. 2001).

The tests showed that the biological process was highly efficient in removing formaldehyde. The supernatant's turbid appearance at high concentrations (750 and 1,000 mg/L) can be explained by the lack of resistance of some microorganisms in the activated sludge to the high toxicity of formaldehyde, leading to cell lysis. A previous study indicated that the toxicity of formaldehyde to microorganisms depends on its concentration, and the 50% inhibitory concentration of formaldehyde was about 300 mg/L (Lu and Hegemann 1998). TOC monitoring revealed that at high formaldehyde concentration, TOC level decreased slowly. The remaining TOC originated from organic compounds of microbial cells. These

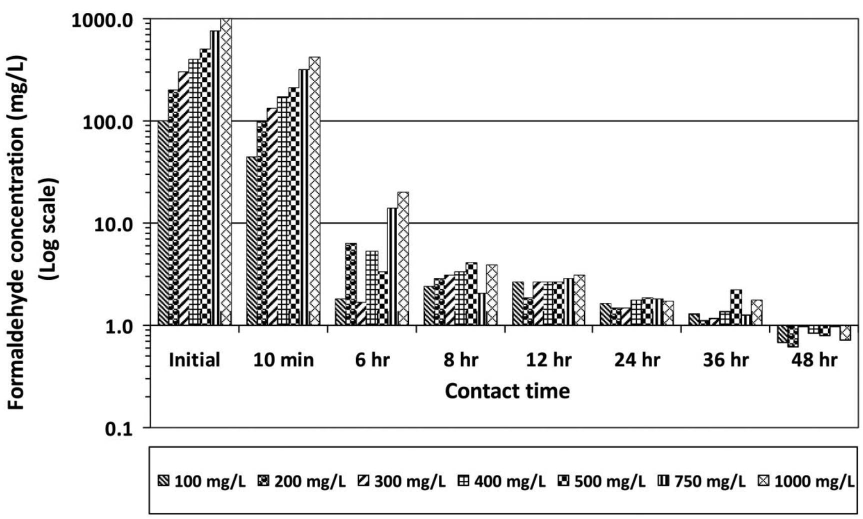

Fig. 2. Formaldehyde concentrations remaining in effluent from batch tests at different initial concentrations and contact times

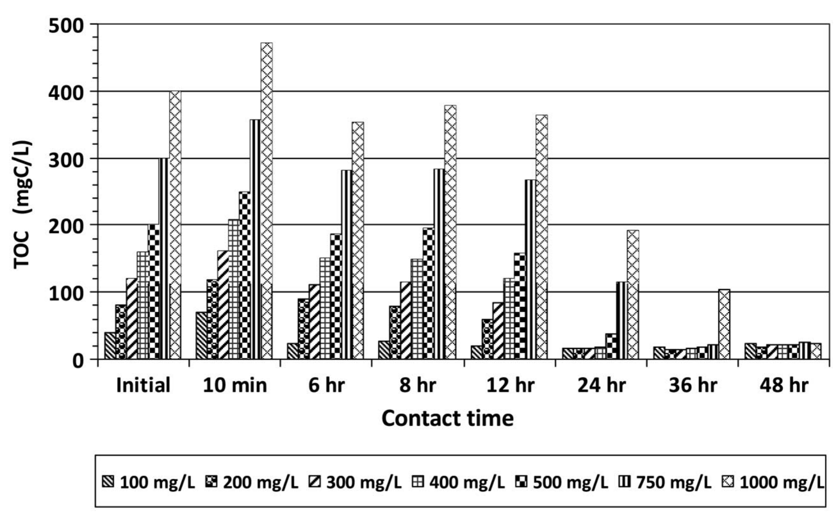

Fig. 3. Total organic carbon concentration in effluent from batch tests at different initial formaldehyde concentrations and contact times

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so-called soluble microbial products (SMPs) produced high TOC levels and turbidity in the effluent (Grady et al. 1999). The SMPs were used as substrates for other microorganisms, so that TOC levels finally decreased at contact times longer than 24 h, as shown in Fig. 3.

Effect of pH on Formaldehyde Removal

The pH of formaldehyde-containing wastewater was varied at 3, 5, 7, and 9. The initial formaldehyde concentration was 1,000 mg/L, and the initial MLSS concentration, approximately 1,000 mg/L. As shown in Fig. 4, formaldehyde was efficiently removed at pH 7, which is a typical pH for biological processes. At this pH, formaldehyde was decreased to below 1 mg/L within 8 h of contact time. However, at pH 5, biodegradation of formaldehyde was still occurring, even more slowly than at pH 7. Therefore, the optimum pH range for formaldehyde removal is 5–7.

Effect of Mixed Liquor Suspended Solids on Formaldehyde Removal

Various MLSS concentrations were used: 500, 1,000, 2,000, and 4,000 mg/L. Initial formaldehyde concentration was approximately 1,000 mg/L, and the pH of the solution was maintained at 7 using phosphate buffer. The results showed that MLSS concentration affects formaldehyde removal efficiency. The higher the MLSS content, the higher the efficiency with which formaldehyde was removed, as illustrated in Fig. 5. At MLSS concentrations of 500 and 1,000 mg/L, over an 8-h period, formaldehyde concentration decreased gradually. The final concentration of formaldehyde in the effluent was still greater than 1 mg/L. As MLSS concentration increased, formaldehyde was removed more rapidly. At MLSS concentrations of 2,000 and 4,000 mg/L, formaldehyde concentration decreased from 1,000 to 1 mg/L within 2 h, and the overall efficiency of formaldehyde removal was as high as 99.9%.

Performance of Membrane Bioreactor in Formaldehyde Removal

The MBR was operated in continuous mode with a flow rate of 1.2 L/h and HRT of 10 h. Acclimated sludge was introduced into the bioreactor with an initial MLSS concentration of 2,000 mg/L. When the system was started up, the concentration of formaldehyde in the influent was gradually increased from 100 to 500 mg/L and was maintained at approximately 500 mg/L. Fig. 6 shows that the

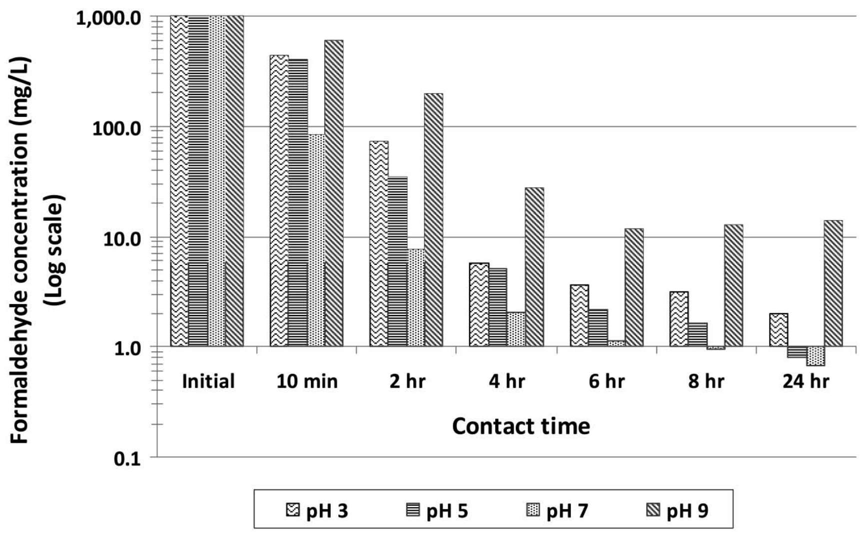

Fig. 4. Effect of pH on formaldehyde removal

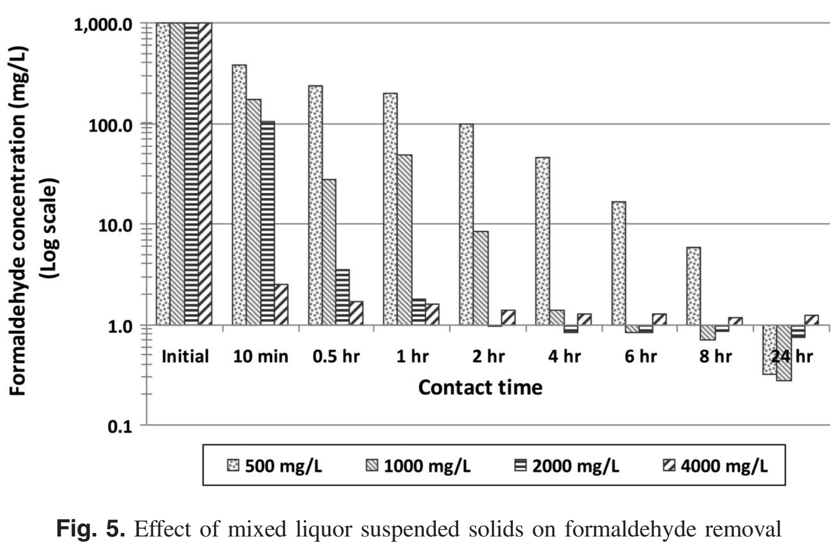

Fig. 5. Effect of mixed liquor suspended solids on formaldehyde removal

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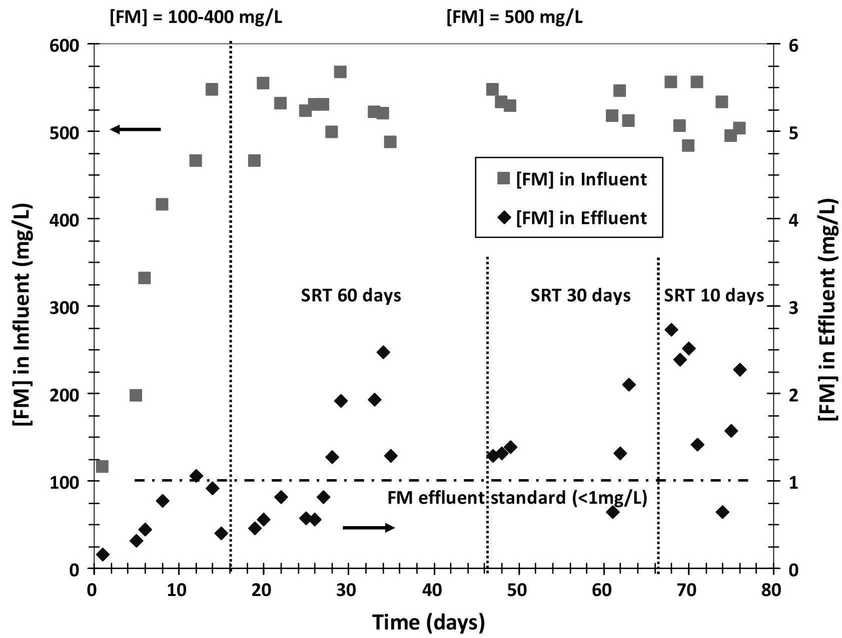

Fig. 6. Performance of membrane bioreactor in formaldehyde removal

MBR reduced the concentration of formaldehyde in the effluent from 526 ± 30 to 1.39 ± 0.73 mg/L, corresponding to a removal efficiency of $99.73 \pm 0.14\%$. This result was similar to that reported by Eiroa et. al. (2005). It was also found that the MBR system was more efficient at an SRT of 60 days than at SRTs of 30 and 10 days, possibly because of the higher MLSS concentration.

Fig. 7 illustrates the concentration of TOC, which represents the organic compounds in solution. As formaldehyde was the sole carbon source for the MBR, the TOC of the influent represented formaldehyde only. Theoretically, 1 mg/L formaldehyde contains 0.4 mg/L TOC, so the formaldehyde remaining in the MBR, 1.39 ± 0.73 mg/L, accounted for the TOC concentration of 0.56 mg/L. However, the effluent concentration of TOC in the MBR was stable at an average of 5.16 ± 1.20 mg/L, indicating that the organic compounds remaining in bulk solution comprised not only formaldehyde but also SMPs. These compounds might be intermediates or products generated by substrate metabolism during biomass growth and/or organics released from biomass decay (Shin and Kang 2003; Jarusutthirak and Amy 2006).

As biodegradation of formaldehyde was confirmed, the MBR performed a specific function, retention of formaldehyde-acclimated biosolids in the bioreactor, resulting in the stability of the system. Additionally, the MBR performed well in TSS removal, with an efficiency close to 100%. The TSS concentration in the effluent was found to be essentially zero, and the effluent appeared clear. Therefore, the effluent of the MBR can be further used as raw water for reuse or recycling.

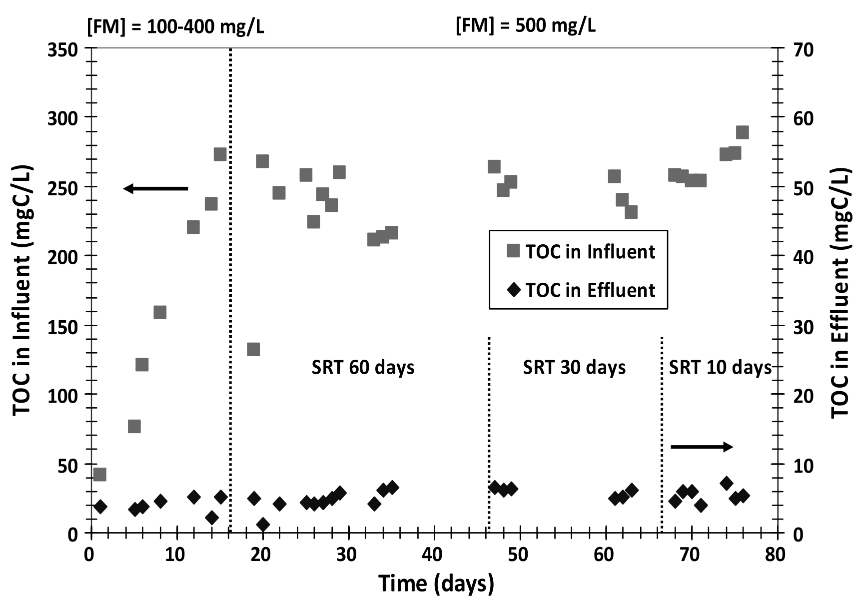

Fig. 7. Performance of membrane bioreactor in removal of total organic carbon

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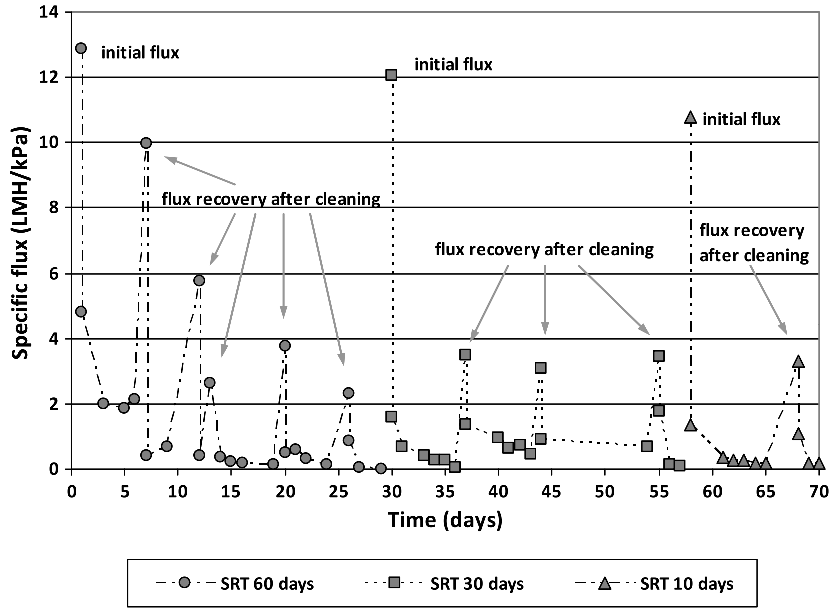

Fig. 8. Flux decline during membrane bioreactor operation at different solid retention times and flux recovery after membrane cleaning

Effect of Solid Retention Time on Membrane Bioreactor Performance

The effect of SRT on formaldehyde removal in the MBR was investigated. It was found that at SRTs of 60, 30, and 10 days, the MBR removed formaldehyde with efficiencies of 99.79 ± 0.13, 99.75 ± 0.09, and $99.62 \pm 0.15\%$, respectively. According to ANOVA, mean formaldehyde removal efficiencies at SRTs of 60 and 30 days significantly differed from that at an SRT of 10 days with 95% confidence. Formaldehyde removal was more efficient at the longer SRTs (60 and 30 days) than at the shortest SRT (10 days).

Flux Decline during Membrane Bioreactor Operation

Specific flux of membranes during MBR operation at different SRTs was monitored. Fig. 8 shows specific flux changes and the cleaning cycle of the membranes. During a typical cycle of operation (5–7 days), suspended solids accumulated on the membrane surface and specific flux decreased. The results revealed similar trends in flux decline at the different SRTs, in agreement with previous studies reporting that MLSS concentration, floc size, extracellular polymeric substances (EPS), and colloidal and soluble organic substances are important contributors to membrane fouling (Bouhabila et al. 2001; Zoh and Stenstrom 2002; Meng et al. 2006; Rosenberger et al. 2006; Huang and Wu 2008). The suspended and/or soluble solids attached to the membrane surface or clogging the membrane pores caused an increase in membrane resistance (Huang and Wu 2008).

At the end of the cycle, increases in specific flux were the result of membrane cleaning. At an SRT of 60 days, rinsing with tap water and chemical cleaning restored 77.5% of original flux after the first cycle. Flux recovery continuously decreased to 44.6, 29.3, and 10.1%, correspondingly, for the next three cycles. Membrane flux recovery after a cycle at SRTs of 30 and 10 days was approximately 28.9 and 30.7%, respectively. It was found that at longer sludge age (SRT = 60 days), bioflocs were very strong because there were more filamentous organisms. Therefore, bacteria tended to stick together in bioflocs, fouling the membrane through caking (Zoh and Stenstrom 2002; Meng et al. 2006). The cake could be removed easily by flushing with tap water for 15 min, leading to high flux recovery. On the other hand, at shorter sludge age, with less MLSS, dispersed bacteria and other microorganisms might clog membrane pores, blocking them, or might adsorb to the membrane surface, forming a gel. This phenomenon made cleaning difficult that flux recovery slowed down within a short period of operation.

Conclusions

Results from batch tests showed the potential of formaldehyde biodegradation using acclimated sludge. Formaldehyde concentration was decreased to below 1 mg/L, corresponding to a removal efficiency of 99.9%, at a contact time of 24 h. At high formaldehyde concentrations (750 and 1,000 mg/L), the effluent appeared highly turbid as a result of the high concentration of TSS in the effluent. The optimum pH range for formaldehyde removal was 5–7. In this pH range, formaldehyde removal efficiency was 99.9% over an 8-h period. The greater the MLSS concentration, the higher was the efficiency of formaldehyde removal. An increase in MLSS enhanced the efficiency of formaldehyde removal at shorter contact time. At MLSS concentrations in the range 2,000–4,000 mg/L, formaldehyde concentration decreased from 1,000 mg/L to approximately 1 mg/L within 2 h, for an overall efficiency of formaldehyde removal as high as 99.9%.

Results from this study confirm the potential for use of MBR in removal of formaldehyde. The MBR exhibited the advantages of retention of acclimated sludge and high quality of effluent. It reduced formaldehyde concentration from 526 ± 30 to 1.39 ± 0.73 mg/L, corresponding to a removal efficiency of $99.73 \pm 0.14\%$. Longer SRTs (60 and 30 days) were associated with higher removal efficiency than short SRT (10 days). The quality of permeate was appropriate for water reuse or reclamation. However, the limitations of the MBR include membrane fouling and declining flux, caused by accumulation of MLSS on the membrane surface. This study indicated that the specific flux of the permeate decreased continuously during MBR operation. SRT did not affect declining flux, but did affect flux recovery after cleaning. A long SRT (60 days) led to greater flux recovery than shorter SRTs (30 and 10 days).

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