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Biological degradation of dissolved organic carbons and ammonia oxidation by biological activated carbon in PAC and membrane applied process

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ABSTRACT

Membrane filtration process has been developed to overcome the weakness of conventional water treatment process for the removal of particulates and some pathogens such as cryptosporidium and giardia. Although suspended particulate, including pathogens and bacteria, can be effectively removed by membrane, the trace organics and soluble matters are hard to remove. To overcome this limitation, membrane filtration is required to be associated with other technologies such as adsorption and oxidation. PAC membrane retrofitting (PMR) process is one of the new processes for removing not only particulate but also soluble matters. PMR process consists of a coarse powder-activated carbon (C-PAC) contactor and a subsequent submerged membrane tank. Powdered activated carbon (PAC) with high concentrations of 30,000 mg L^{-1} is suspended as a slurry blanket in the PAC contactor and soluble matters are removed by the slurry blanket. Most particulates are separated by the membrane. Membrane module used in this study was ZeeWeed® 500℃ with 603 m^2 of effective surface area. The membrane is made of polyvinylidene fluoride (PVDF) with nominal pore size of 0.04 μm. Operating flux of membrane varied from 21 L m^{-2} hr^{-1} (LMH) to 42 LMH. Recovery rate of the process was maintained at 99.5%. The purpose of this study was to investigate the biological removal performance of PMR process. Dissolved organic carbon (DOC) concentration of influent and effluent of each unit process were measured. Ammonia step-feeding tests were performed to evaluate the oxidation of ammonia by microorganisms attached on the PAC. Also, the number of bacteria on PAC surface was counted to estimate the build-up of bio-film. After the PAC in the PAC contactor was exhausted and turned into biological activated carbon (BAC), the average bio-film density of BAC was 10^6 cfu g^{-1}. Ninety-four percentage of the bacteria were attached on the BAC in PAC slurry blanket, and the rest was suspended. When the contact time of raw water through the slurry blanket was prolonged from 23 to 45 min, average NH_4^+-N removal was increased from 76 to 97% and

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average DOC removal was raised from 7 to 17%. Ammonia and DOC were consistently and effectively removed by the BAC in the PMR process.

Keywords: Membrane; Biological activated carbon (BAC); PMR process; Dissolved organic carbon (DOC)

1. Introduction

Recently, the removal of trace organics, such as chlorination by-products, endocrine disruptors, pharmaceuticals and taste and odour-causing compounds, from the drinking water has become a significant issue for water authorities due to the enforcement of stricter regulations and consumer's demand for better quality water.

Membrane filtration processes have been developed to overcome the weakness of conventional water treatment process in the removal of particulates. Membrane processes remove particulate matters and some pathogens such as *Cryptosporidium and Giardia* much better than conventional media filters do [1,2]. Although suspended particulates including pathogens and bacteria can be absolutely removed by membrane, trace organic compounds and soluble matters are still hard to remove [3,4].

To overcome this limitation, membrane filtration as well as media filtration is often associated with other technologies such as adsorption and oxidation. As a result, a few hybrid processes combining adsorption and membrane filtration have been proposed to absolutely exclude the suspended particles from water (via membrane filtration) and effectively remove soluble organics (via adsorption by activated carbon). Especially, a combination of powdered activated carbon (PAC) or granular-activated carbon (GAC) and submerged membrane has been applied to treat water [4–6].

PAC is intermittently applied before a membrane filtration process [7–9]. When PAC is injected ahead of a submerged membrane tank, it accumulates in the membrane system such that all the adsorption capacity of the carbon is utilized for removing soluble organics. However, effects of PAC on membrane fouling have not been well established when the system was operated with high concentration of PAC in the membrane tank [6,9–11].

The enhanced trace organic compounds removal through PAC addition was reported in drinking water treatment combined with membrane process [6,12–14]. Some researchers reported the removal of organic substances and ammonium by employing biological activated carbon (BAC) converted from GAC [15–17]. Moreover, NOM was absorbed onto the bio-film and then was slowly biologically degraded [18].

A few previous studies on the DOC removal by BAC are summarized in Table 1. Traditionally, the formation of bio-film and its removal of organic matter were reported in the GAC adsorption process. The DOC removal efficiencies of bio-film, which have been reported in the literature, vary from 13% to 72%.

However, the application of the PAC is often considered not to be economical because PAC, once added to the system without recycle, is easily discharged from the system along with the concentrate of the membrane process; consequently, it is not kept in the system long enough to exhaust the adsorption capacity of the PAC. Installation of GAC adsorption tower after membrane filtration, potentially results in the leaks of microorganisms which may grow on the GAC surface into the final treated water [25].

In this study, an innovative hybrid system consisting of a PAC contactor and a membrane filtration tank was developed to treat surface water. Large amount of PAC having a size of 90–180 μm was put into the PAC contactor and kept in suspension to maintain PAC slurry blanket at a certain height in the contactor. PAC particles in suspension can adsorb trace organic contaminants in raw water. Also, microorganisms introduced into the system with the raw water can attach on the surface of PAC particles. Eventually, they grow up to form a natural bio-film, which degrade organics presented in the raw water. Owing to the bio-film formation on PAC particles, the PAC contactor could still maintain good organic removal even after the adsorption capacity of PAC is exhausted; it is main advantage of the PMR process [26–32].

The main objectives of the current research were to evaluate the bio-film characteristics on the PAC particles and to evaluate ammonia oxidation by the bio-film. DOC removal efficiency of the PAC contactor was also evaluated with different contact time of raw water through the slurry blanket gave different slurry depths.

Table 1
Typical DOC removals by biological activated carbon (BAC) in the literature

Water source	DOC (mg L^{-1})	Media	O_3 dose	Removal (%)	Reference
Plonia river, Poland	7.8–11.6[a]	GAC	1.64 mg O_3/mg TOC	39	Emelko et al. [19]
Grand river, USA	5–7	GAC	NA	13–23	Li et al. [16]
Miyun reservoir, China	4.9–7.3	GAC	3 mg L^{-1}	33.4	Xu et al. [17]
Huangpu river, China	5.2–7.7	GAC	2.0–2.5 mg L^{-1}	31	Hozalski et al. [20]
Omerli reservoir, Turkey	2.9–4.9	GAC	No ozonation	47–72	Yapsakli et al. [21]
SWWE from BWTP[b], Australia	9.5–10.6	GAC	No ozonation	21–31	Aryal et al. [22]
Nakdong River, Korea	1.5–2.8	PAC	No ozonation	20–40	Seo et al. [5]
Han River, Korea	1.2–3.0	GAC	No ozonation	17–20	K-water[c] [23]
Han River, Korea	1.2–3.0	GAC	1 mg L^{-1}	26–30	K-water[c] [23]
Han River, Korea	NA	GAC	No ozonation	36–47	WRI, Seoul[d] [24]

[a]In terms of TOC.
[b]Secondary wastewater effluent from Beenyup wastewater treatment plant.
[c]Korea Water Resources Corporation.
[d]Waterworks Research Institute, Seoul Metropolitan Government.

2. Materials and methods

2.1. Experimental set-up

A pilot scale plant was installed in a drinking water treatment plant in Namyangju-City, Korea. Raw water is supplied to the plant from Paldang Reservoir without pre-treatment. The pilot plant consisted of a PAC contactor with PAC slurry blanket and a submerged membrane tank with a ZeeWeed® 500C membrane module as shown in Fig. 1.

The cylindrical PAC contactor has the effective volume of 16 m^3 with a diameter of 3,100 mm and a height of 2,100 mm. A mechanical mixer was installed in centre of the contactor and continuously stirred to keep PAC particles in suspension. The effluent of the contactor flowed to the membrane tank over weirs at the top of the contactor.

2.2. Heterotrophic plate count (HPC)

The heterotrophic plate count is a standard method to estimate the number of viable heterotrophic bacteria in water. There is no single medium for supporting all heterotrophs in water. Nevertheless,

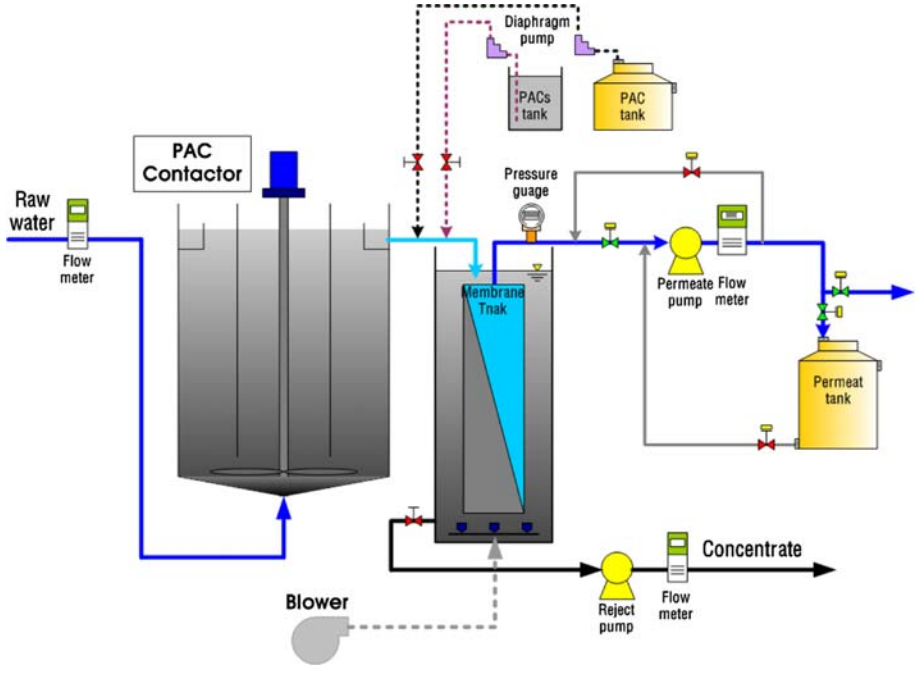

Fig. 1. Experimental set-up of PMR plant.

heterotrophic plate count provides a good measure of process efficiency of a water treatment process in eliminating microbial cells.

2.2.1. Media preparation

NWRI (National Water Research Institute) agar, also known as heterotrophic plate count agar (HPCA) was used. The medium was made by adding the following substances in deionized water of 1 L:3.0 g peptone, 0.5 g soluble casein, 0.2 g K_2HPO_4, 0.05 g $MgSO_4$, 0.001 g $FeCl_3$ and 15.0 g agar. The pH of the medium was adjusted to 7.2. Then, the medium was autoclaved for 15 min at 121°C and 1.03 bar.

2.2.2. Sample preparation

Samples collected from PMR pilot plant were stored at 4°C until they were analysed; albeit all the samples analyzed within 24 h. The samples were fractionated and labelled as follows: a. **supernatant**—the liquid fraction after PAC settled down, b. **mixture**—the mixture of supernatant and PAC, and c. **sonicated mixture**—the mixture of supernatant and PAC treated with ultrasonication. About 200 mL of each well-mixed fraction was collected in separate 300 mL glass Erlenmeyer flasks. Ultrasonication treatment was employed using Bransonic® 1,510 (Branson, USA) ultrasonic cleaner (70 W, 42 kHz) for 10 min.

2.2.3. Pour plating

About 1 mL aliquot from each dilution was poured on a disposable Petri dish (100-mm diameter); all samples were prepared in triplicate. Samples were mixed well prior inoculation to ensure evenness of size distribution. Each plate was poured with approximately 12 mL of NWRI agar. The NWRI medium was thoroughly mixed with the sample by rotating the Petri dish in opposite directions. All the plates were allowed to be solidified prior incubation. All inoculated plates were inverted, put in a plastic bag, and incubated at 35°C for 48 h. Well-developed colonies were counted following the Standard Methods [33].

2.3. Ammonia step-feeding tests

After the adsorption capacity of the PAC added into the PAC contactor in the beginning was exhausted, organic matter removal by microorganisms on the BAC was evaluated. The prepared stock solution was continuously injected into the raw water for more than 24 h, to keep the ammonia concentration of the feed water 0.5 mg L^{-1} and 1.0 mg L^{-1}.

Influent and effluent of the PAC contactor and membrane permeate were collected every 3 h during the test. The samples were filtered with 0.45 μm syringe filter (MACHEREY-NAGEL GmbH & Co. KG, Germany) to remove particulates such as PAC. The concentrations of ammonium ion were measured by ion chromatography (761 compact IC, Metrohm AG, Switzerland) equipped with a Metrosep C 4 cation column (Metrohm AG, Switzerland).

Operating conditions of the PMR process were summarized in Table 2.

2.4. Evaluation of DOC removal efficiency

To evaluate the performance of the PMR process, DOC concentrations of influent and effluent of the PAC contactor and membrane permeate were measured once a day. Water samples were filtered using syringe filters (0.45 μm) before the analysis, and their DOC concentrations were measured with a total organic carbon analyzer (Sievers* 900, GE Analytical Instruments, USA).

Especially, when DOC removal efficiency of the BAC was evaluated, coagulant and fresh finer PAC (with average diameter ranged from 23 to 42 μm) were not added.

3. Results and discussion

3.1. Heterotrophic plate count

The number of heterotrophic bacteria samples collected from the PAC contactor was analysed; the water samples were collected at different depths. The number of heterotrophic bacteria slightly increased with depth as shown in Fig. 2. However, taking into account, the error bars, cell counts between samples collected at 1.0 m and at 1.9 m was not significantly

Table 2
Operating conditions of PMR process during the ammonia step-feeding test

Operation capacity ($m^3\,day^{-1}$)	HRT* of the PAC contactor (min)	Height of the slurry blanket (m)	Slurry blanket contact time of raw water (min)
550.0	41	1.3	23
412.5	54	1.3	30
		1.5	38
275.0	81	1.3	45

*HRT: Hydraulic retention time.

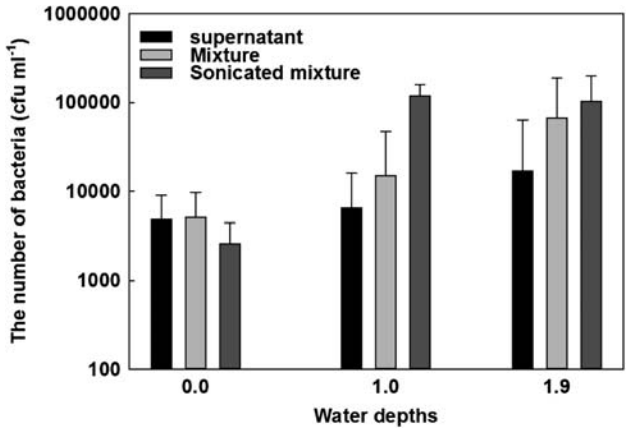

Fig. 2. Average number of bacteria with water depth in the PAC contactor.

different due to the similar concentration of the PAC slurry in the PAC contactor. The PAC concentration of the PAC slurry taken from at 1.0 m and 1.9 m ranged from 40,000 mg L^{-1} to 55,000 mg L^{-1}, while the one of the sample collected from just below the water surface of the contactor was well under 100 mg L^{-1}. The number of bacteria of surface water sample was also relatively low. This result showed that the bacterial population of the bio-film formed on the surface of PAC particles was proportional to the concentration of PAC.

Table 3 summarizes bacterial populations on BAC particle present in GAC filtration units in drinking water treatment plant. The bio-film density of PAC particles in the PMR process were 3×10^6 cfu g^{-1}. Bio-film densities reported in the literature ranged from 10^4 to 10^{11} cfu g^{-1}. The bio-film formed on PAC particles in the PAC contactor could grow up to the similar level of bio-film in GAC filtration process which has good biodegradation efficiency.

Bacteria in the supernatant, which is the liquid fraction after PAC of the sample settled down, were considered to be originated from the raw water. On the other hand, bacteria in "sonicated mixture" might represent the sum of suspended bacteria in the contactor and all the bacteria detached from the PAC particles. Therefore, the difference between sonicated mixture and supernatant can be considered as the number of bacteria detached from the PAC surface. The ratio of attached bacteria can be written as follows:

$$\text{The ratio of attached bacteria } (\%) = \frac{(\text{sonicated mixture} - \text{supernatant})}{\text{sonicated mixture}} \times 100$$

The ratio of attached bacteria in samples collected at 1.0 m was founded to be 89 to 99% and the average ratio was 94.1% as shown in Fig. 3. This result indicated that the removal of organic substances by bio-film could be expected even after the adsorption capacity of the PAC is exhausted.

3.2. Evaluation of ammonia oxidation in the PAC contactor

Typically, ammonia in the water could be removed via biological oxidation or air stripping. The reduction of ammonia (as NH_4^+ in the PAC contactor was attributed to the presence of nitrifiers on the PAC particles. During the step-feeding test, the influent and effluent NH_4^+-N concentration profiles along with removal efficiencies of each unit process are presented in Fig. 4. The ammonia removal efficiency ranged from 63% to 93% in the PAC contactor, which showed that the most nitrification took place in the PAC contactor.

Table 3
Attached bio-film density on BAC

Media	Bio-film density		Reference
	(cfu ml^{-1})	(cfu g^{-1})*	
PAC	10^5	3×10^6	This research
GAC	–	5×10^7	Camper et al. [34]
GAC	–	10^4	Camper et al. [35]
GAC	–	10^7	LeChavallier et al. [36]
GAC	$10^3 \sim 10^5$	$10^7 \sim 10^{11}$	Pernitsky et al. [37]
GAC	10^3	–	Waterworks Research Institute, Seoul Metropolitan Government [24]
GAC	–	10^6–10^8	Korea Water Resources Corporation [23]

*(cfu mL^{-1}) divided by PAC concentration.

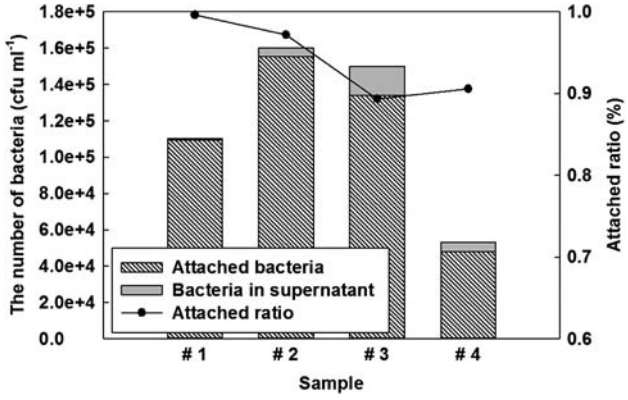

Fig. 3. Number of bacteria and attached ratio.

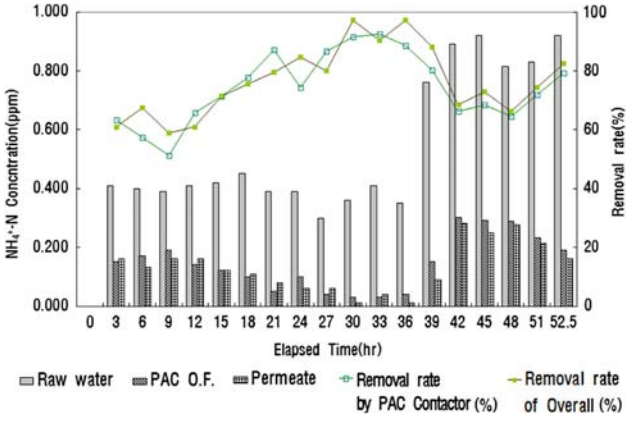

Fig. 4. Variation of the NH_4^+-N concentration and removal rate.

Similar NH_4^+ removal efficiency of bio-film in GAC filtration have been reported in the literature. Yapsakli et al. [38] reported that more than 97% of NH_4^+ was removed in their experiments with a GAC column. Andersson et al. [15] and Laurent et al. [39] reported nitrification efficiency higher than 90% in their full-scale GAC filters.

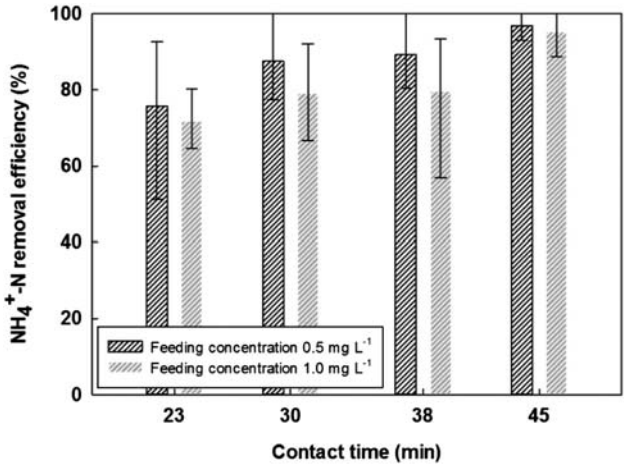

Fig. 6. Average removal efficiencies by C-PAC contactor with contact time.

After 35 h of operation with step feeding of ammonia concentration of 0.5 mg L^{-1}, the influent NH_4^+ concentration was raised to 1.0 mg L^{-1} to evaluate the nitrifiers' adaptability for a sudden change in inflow water quality. Even after the sudden change, the system's performance was not affected much. Although the NH_4^+ removal efficiency of the system was initially decreased to 70% due to a sharp increase in inflow concentration, the removal efficiency was recovered to about 80% after 10 h.

Finally, the impact of contact time of raw water through the slurry blanket of the PAC contactor was evaluated for better understanding of nitrification in the PMR process. When the contact time of raw water

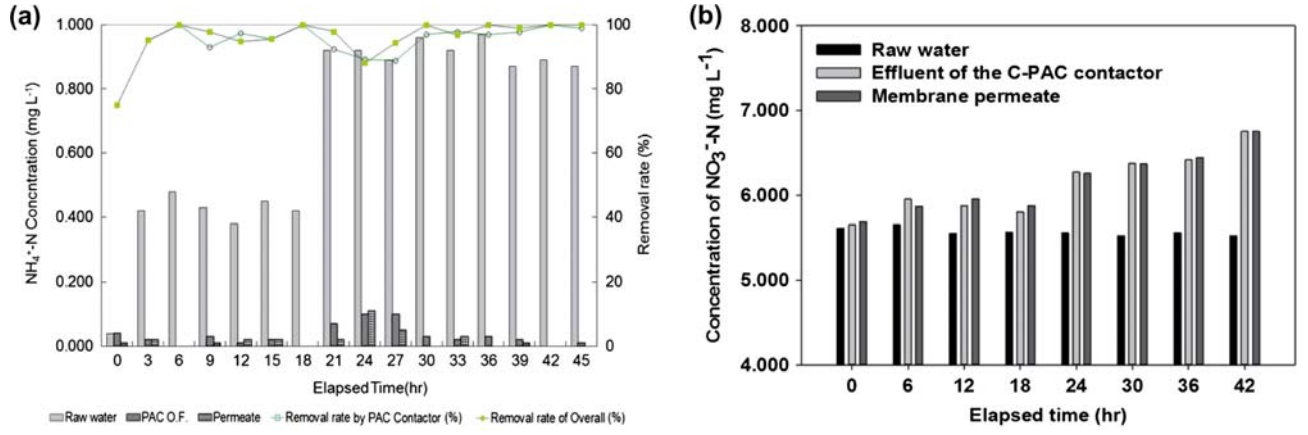

Fig. 5. Variation of the NH_4^+-N and NO_3-N concentration and removal rate (the contact time: 45 min).

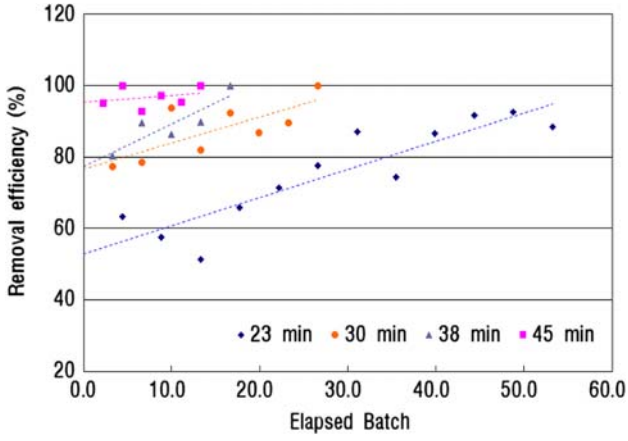

Fig. 7. Variation of NH_4^+-N removal efficiencies with elapsed batch at different contact time.

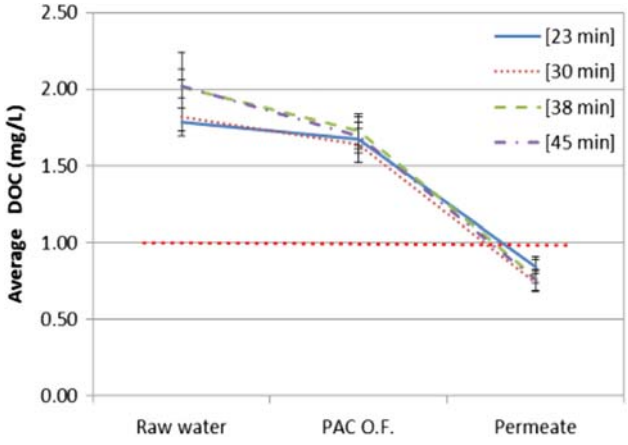

Fig. 8. Average concentration of DOC with contact time.

with C-PAC slurry was given as 45 min in the C-PAC contactor, the concentration of NH_4^+-N and its removal rate was presented with time in Figs. 4 and 5(a). More than 90% of ammonia was removed in the PMR process and most that was removed by bio-film in the C-PAC contactor. Concentration of nitrate was also measured to confirm the nitrification. Nitrate concentrations of raw water and effluent from each unit process were shown in Figs. 4 and 5(b) with elapsed time. Concentration of nitrate increased after contact with C-PAC slurry blanket but it was not changed in the membrane tank. These observations indicate that the most nitrification took place in the C-PAC contactor. Converted nitrate in the C-PAC contactor increased after the elapsed time of 24 h due to the change of feeding concentration of NH_4^+-N.

Average removal efficiencies are summarized in Fig. 6. The NH_4^+ concentration of the feed was adjusted to 0.5 mg L^{-1} by adding appropriate NH_4Cl stock solution. The contact time varied from 23 to 45 min. The average removal efficiency was proportional to the length of the contact time. The removal efficiency was over 95%, when the contact time was 45 min.

Fig. 7 shows removal efficiency at different elapsed batch, which is the volume of PAC contactor. The NH_4^+ removal efficiency reached its maximum at a faster rate as the contact time increased. When the contact time was 45 min, the maximum efficiency was obtained in less than five batches. However, the maximum efficiency was obtained in over 50 batches for the contact time of 23 min.

It should be noted that the removal efficiency eventually could reach at 90% even at lower contact times, as the system was operated longer. It was assumed that more nitrifiers grew up on the PAC particles as the influent NH_4^+ concentration is increased.

3.3. Evaluation of DOC removal efficiency

Fig. 8 shows the DOC concentrations of influent and effluent of PAC contactor and the membrane permeate in the PMR process. Even though DOC concentrations of the influent was in the range from 1.69 to 2.24 mg L^{-1}, the permeate DOC was always below 1 mg L^{-1} during the operation. The high removal of DOC in the membrane tank was due to the addition of coagulant (Poly Aluminium chloride silicate) and fresh PAC to ahead of the membrane tank.

When the contact time of the raw water was 23 min, average DOC concentration of the raw water and overflow from the PAC contactor was 1.78 mg L^{-1} and 1.67 mg L^{-1}, respectively showing about 6.58% removal efficiency. The efficiency of the PAC contactor increased from 9.23% to 16.6% as the contact time increased up to 45 min as shown in Table 4. The result indicates that the contact time would be a key

Table 4
DOC removal efficiency of PMR process with the contact time

Contact time (min)	Removal rate by the PAC contactor (%)	Removal rate by the membrane tank (%)	Overall removal rate (%)
23	6.6	46.0	52.6
30	9.2	49.9	59.1
38	14.2	47.3	61.5
45	16.6	46.0	62.6

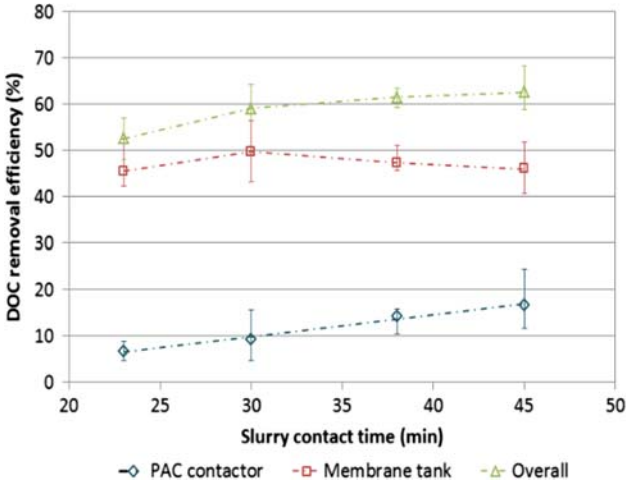

Fig. 9. DOC removal efficiencies with contact time.

operational parameter for the PAC contactor in terms of DOC removal.

As shown in Fig. 9, DOC removal efficiency of the PAC contactor linearly increased with the contact time. However, the removal efficiency in the membrane tank was not increased at the contact time longer than 30 min even though the efficiency was much higher than that of the PAC contactor. Since the coagulant and fresh PAC was added to the membrane tank, organic matters are rapidly adsorbed on the PAC and removed by the coagulation. The DOC removal efficiency of more than 60% could be achieved in the PMR process.

4. Conclusions

In this study, it was confirmed that C-PAC was converted to BAC in the PAC contactor. In addition, DOC and ammonia removal performance of a pilot scale PMR process was evaluated. From the study, the following conclusions were derived:

- After the adsorption capacity of PAC particles in the PAC contactor was exhausted, the number of attached bacteria on PAC particles was 3×10^6 cfu g^{-1} and the average ratio of attached bacteria in the slurry blanket was estimated to be 94.1%. The PAC in the contactor was converted to BAC and the removal of organic matters could be possible by bio-film formed on the surface of the particles.
- The NH_4^+-N removal rate of the contactor became faster as the contact time increased from 23 to 45 min. In addition, the removal efficiency could reach to more than 90% as the contact time increased.
- DOC concentration of the permeate could be maintained below 1 mg L^{-1} with addition of coagulant and fresh PAC in the PMR process. The DOC removal efficiency of the bio-film on the PAC particles in the PAC contactor increased from 7 to 17% in proportion to the contact time.

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