Long term operation of high concentration powdered activated carbon membrane bio-reactor for advanced water treatment

G.T. Seo*, C.D. Moon, S.W. Chang** and S.H. Lee****

* Department of Environmental Engineering, Changwon National University, Sarim-dong 9, Changwon, Gyeongnam 641-773, South Korea (E-mail: *gts@sarim.changwon.ac.kr*)

** Korea Water Resources Co. Southern Nakdong River Regional Water Quality Analysis Center, 25 Banlim-dong, Changwon, Gyeongnam, South Korea

Abstract A pilot scale experiment was conducted to evaluate the performance of a membrane bioreactor filled with high concentration powdered activated carbon. This hybrid system has great potential to substitute for existing GAC or O_3/BAC processes in the drinking water treatment train. The system was installed at a water treatment plant located downstream of the Nakdong river basin, Korea. Effluent of rapid sand filter was used as influent of the system which consists of PAC bio-reactor, submerged MF membrane module and air supply facility. PAC concentration of 20 g/L was maintained at the beginning of the experiment and it was increased to 40 g/L. The PAC has not been changed during the operational periods. The membrane was a hollow fiber type with pore sizes of 0.1 and 0.4 μm. It was apparent that the high PAC concentration could prevent membrane fouling. 40 g/L PAC was more effective to reduce the filtration resistance than 20 g/L. At the flux of 0.36 m/d, TMP was maintained less than 40 kPa for about 3 months by intermittent suction type operation (12 min suction/3 min idling). Adsorption was the dominant role to remove DOC at the initial operational period. However the biological effect was gradually increased after around 3 months operation. Constant DOC removal could be maintained at about 40% without any trouble and then a tremendous reduction of DBPs (HAA_5 and THM) higher than 85% was achieved. Full nitrification was observed at the controlled influent ammonia nitrogen concentration of 3 and 7 mg/L. pH was an important parameter to keep stable ammonia oxidation. From almost two years of operation, it is clear that the PAC membrane bioreactor is highly applicable for advanced water treatment under the recent situation of more stringent DBPs regulation in Korea.

Keywords Adsorptions; disinfection byproducts; drinking water; membrane bioreactor; microfiltration; powdered activated carbon

Introduction

Increased organic contamination of the water supply source has led to the realization that conventional water treatment processes can no longer produce safe drinking water. Especially disinfection by products (DBPs) in drinking water have been a hot issue during the last decade in Korea and water supply authorities began to establish more stringent DBPs regulation. Consequently some advanced treatment processes are necessarily required to meet the new standards. Granular activated carbon (GAC) or ozone/biological activated carbon (BAC) had been commonly used for the purpose.

Recently a powdered activated carbon (PAC) coupled membrane system has been introduced as an innovative process for water treatment. The PAC/UF system was developed and used in large scale treatment plants to remove organic micropollutants showing competitiveness with conventional adsorption process (Anselme *et al*., 1997; Baudin *et al*., 1997). The PAC/UF process has the function of adsorption of organics by activated carbon particles that are separated from the product water by the membrane. The overall adsorption efficiency of the hybrid process depends on the reactor configuration and its operational condition (Snoeyink *et al*., 2000). There was other research to apply a high

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concentration PAC coupled microfiltration (MF) system and it was identified that the high concentration PAC in the system could produce enhanced water quality in terms of TOC and E_{260}. The high concentration PAC can also prevent organic fouling of the membrane (Kim *et al*., 2000, 2001). Seo *et al*. (2002) observed ammonia oxidation even at low temperature in the high concentration PAC-MF system. They concluded that PAC in the reactor had an important role in the activity of ammonia oxidizing bacteria. Although the system showed good performance, since the test was carried out in the laboratory scale, a pilot scale investigation is required for successful application of the hybrid system to real water treatment. In this study, a long term pilot scale investigation was conducted to evaluate the system performance and proper operational conditions in a water treatment plant which has difficulties in meeting stringent DBPs regulation.

Materials and methods

Experimental apparatus

The pilot plant was installed at a water treatment plant which takes its raw water from downstream of the Nakdong River flowing in the southern part of Korea. The treatment plant adopts conventional coagulation/flocculation, sedimentation, rapid sand filtration and disinfection. The pilot plant of high concentration powdered activated carbon – membrane bioreactor (HCPAC-MBR) was located at the outlet of the rapid sand filter as shown in Figure 1. A certain amount of effluent from the rapid sand filter was introduced into the system as influent. A hollow fiber membrane module was immersed in the reactor with working volume 300 L and was installed directly above the air diffuser (air flow rate of 60 L/min) to prevent solid accumulation on the membrane surface. Two types of membrane were used in terms of pore size (0.1 and 0.4 micron). Intermittent suction was carried out for filtration. Powdered activated carbon (PAC) was introduced into the reactor at a concentration of 20 g/L at the initial stage and it was increased to 40 g/L for subsequent experiments. The influent flow rate was controlled by a level sensor set in the reactor.

Influent characteristics

The effluent of the sand filter was used as the influent of the system and the water quality is shown in Table 1. Since the ammonia concentration was quite low, it was artificially controlled in the range of 1–7 mg/L for the experiment. Besides DOC and UV_{254}, a standard solution of DBPs (HAA_5 and THM) was added to the influent to evaluate their removal in the system. DBPs were analyzed by gas chromatography (Varian 3200 Series). For pretreatment, HAA_5 was extracted using the Modified EPA 552 micro-extraction method with sulfuric acid addition.

Experimental conditions

The experimental conditions of the pilot plant are shown in Table 2 for about two years of operation (June 2001–May 2003). The operational parameters were initial flux,

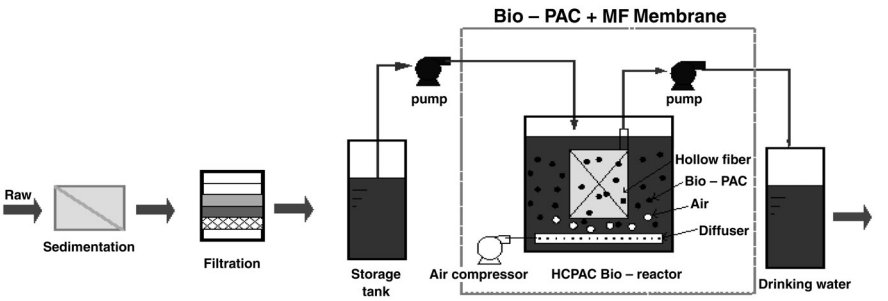

Figure 1 Schematic diagram of HCPAC-MBR in a conventional water treatment process

Table 1 Water qualities of rapid sand filtration effluent

Parameters	Unit	Values
pH	–	6.5–7.2
DOC	mg/L	1.53–2.83
UV_{254}*	1/cm	0.015–0.049
SUVA**	L/mg.m	1.0–1.6
Turbidity	NTU	0.04–0.10
NH_3-N	mg/L	ND-0.02
Alkalinity	mg/L	18–55

*: Absorbance at wavelength 254 nm **: UV_{254}/DOC *100

Table 2 Operating conditions of pilot-plant

Item	Run-1	Run-2	Run-3	Run-4	Run-5
Flux (m/d)		0.72		0.36	
Suction / Idle(min)		90/30		12/3	
PAC conc. (g/L)	20		40		40
Membrane Pore size (μm)		0.1			0.4
Air backwashing	–	–	–	–	+

intermittent suction time interval, PAC concentration in the reactor, membrane pore size, and air backwashing. PAC was replenished for the amount lost during cleaning and replacement of the membrane in Run-4 and Run-5. A membrane with an 0.4 micron pore size was used from experimental Run-4 and air backwashing was introduced at Run-5.

Results and discussion

Adsorption and filtration characteristics of PAC

Prior to the pilot scale experiment, some preliminary tests were carried out to identify adsorption features of various types of PAC and the proper concentration in the reactor. Three types of PAC (lignite, charcoal, coconut shell) were used to estimate the adsorption capacity of organic matter in the influent. Table 3 shows the Freundlich adsorption coefficient for the PAC. The adsorption tests were carried out using KHP (potassium hydrogen phthalate) solution, HAA and THM standard solution. The test results followed the Freundlich equation well for DOC ($r^2 = 0.96$–0.99), HAA ($r^2 = 0.94$–0.95) and THM ($r^2 = 0.95$–0.98), respectively. PAC made of coconut shell had a better adsorption capability for most parameters than other types of activated carbon, which led to their use for the pilot experiment.

Proper PAC concentration in the bioreactor was estimated in terms of transmembrane pressure (TMP) and organic removal. The TMP increase was observed at different dosages of PAC (4 to 80 g/L) in pure water and KHP solution as standard organic matter. The results are shown in Figure 2 and Figure 3. As shown in Figure 2 (a), a higher PAC concentration resulted in the short filtration period at the same TMP, which might be caused by the PAC cake resistance. However, Figure 2 (b) shows that the TMP could be maintained under 40 kPa for extended period of filtration in the reactor with 40 g/L PAC.

Table 3 Freundlich coefficients for various types of PAC

Parameters	Lignite		Charcoal		Coconut shell	
	K	1/*n*	*K*	1/*n*	*K*	1/*n*
OC (mg/L)	5.53	0.7468	12.03	0.6851	11.69	0.3921
THM (μg/L)	253.86	0.6598	87.38	0.7326	373.59	0.6068
HAA (μg/L)	70.41	0.7100	12.31	0.8972	33.99	0.6009

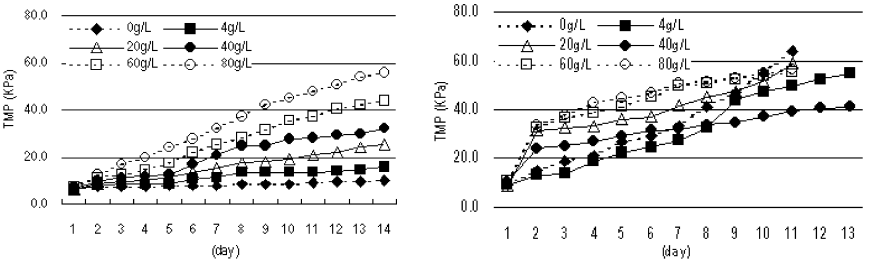

Figure 2 TMP increase by filtration time at various PAC concentrations in pure water (a) and KHP (standard organic matter) solution (b)

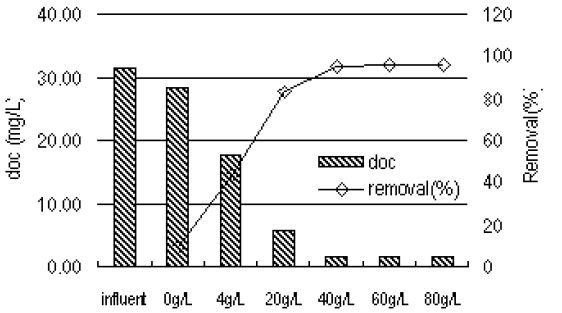

Figure 3 DOC removal at various PAC concentrations in the MF system

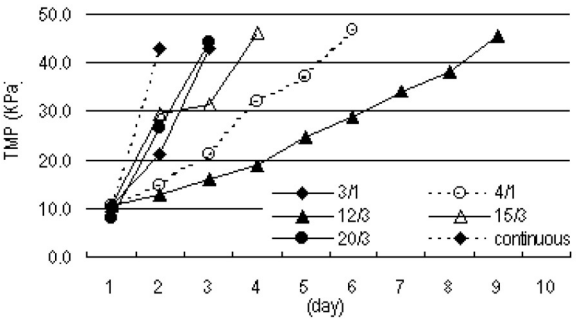

Figure 4 TMP increase by intermittent suction interval (suction/idle time in min.)

DOC removal at different PAC concentrations is depicted in Figure 3. From 40 g/L PAC dosage DOC removal was not improved by the increased PAC concentration. This means that 40 g/L of PAC is enough for organic removal in the bioreactor. Figure 4 shows the TMP increase at different intermittent suction intervals in the MF system. The test conditions are as follows: PAC 40 g/L without sludge inoculation, air supply rate of 10 L/min, water temperature 20 degrees Celsius and initial flux 1.44 m/d. The suction interval of 12 min/3 min was most efficient for stable operation of the filtration system among the conditions.

System performance of HCPAC-MBR

Filtration resistance. Filtration resistance was monitored during the operational period of the pilot plant. The results are shown in Figure 5. Operational flux was set at 0.72 m/d for Run-1 and Run-2. It was reduced to 0.36 m/d from Run-3. Membrane replacement or chemical cleaning was done at TMP of 40 kPa.

Replacement interval of the membrane was about one month at a flux of 0.76 m/d: however it increased to 2–3 months at 0.36 m/d. This result is similar to that obtained by Kim

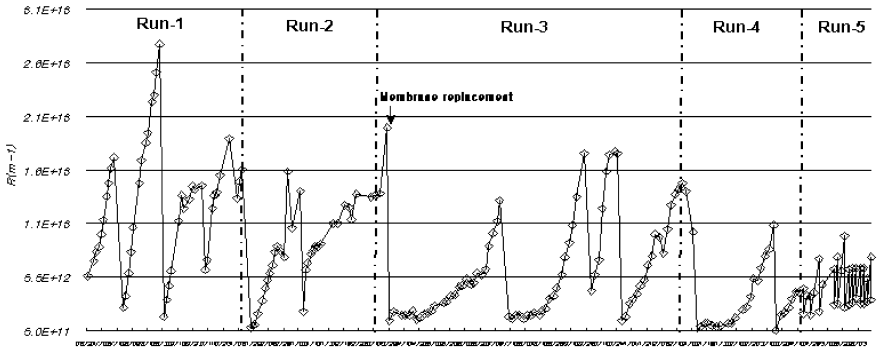

Figure 5 Variations of filtration resistance by operation time of HCPAC-MBR

(2001) from the filtration test at different fluxes, 1.0 m/d and 0.5 m/d for filtration of the effluent of the bio-filter with 40 g/L PAC. He observed also a much faster increase of TMP at the flux of 1.0 m/d. At the flux 0.5 m/d, the TMP could be maintained for 30 days under 20 kPa. Comparing the result of Run-3 (0.1 micron) and Run-4 (0.4 micron), there was no effect of membrane pore size on filtration resistance with high concentration PAC. When air-backwashing was performed once a day in Run-5, the filtration resistance could be maintained stably for an extended period.

Organic removal. Figure 6 shows the removal efficiency of DOC and UV_{254} by the system. Adsorption was the dominant role to remove dissolved organic matter (DOC) during the initial operational period showing the removal of DOC of more than 80%. However it decreased gradually to 30–40%. Although the adsorption effect was decreased by the operational time, organic removal was still maintained above 20% indicating microbial reaction in the system. Actually a bacterial count of 3.4×10^4 cfu/ml was observed after 60 days operation by the heterotrophic plate counting (HPC) method. It could also be estimated that there was some combination between adsorption and biodegradation from the continuous removal efficiency of UV_{254} higher than DOC for the whole experimental period.

DBPs removal. Figure 7 shows the average concentration of DBPs (HAA_5 and THM) in the influent and effluent of HCPAC-MBR. It was comparable with that of the finished water of the conventional treatment process. The removal efficiency of HAA_5 was 92.1% (Inf. 1.0–78.2 ppb average 28.5 ppb, Eff. ND-16.5 average 2.3 ppb) by the system and total THM, 86.1% (Inf. 9.2–57.0 ppb average 28.7 ppb, Eff. ND-9.0 ppb average 4.0 ppb). The removal of DBPs seems to be due to both adsorption and biological degradation in the hybrid system. Consequently the HCPAC-MBR system is excellent in removing DBPs for safe drinking water production.

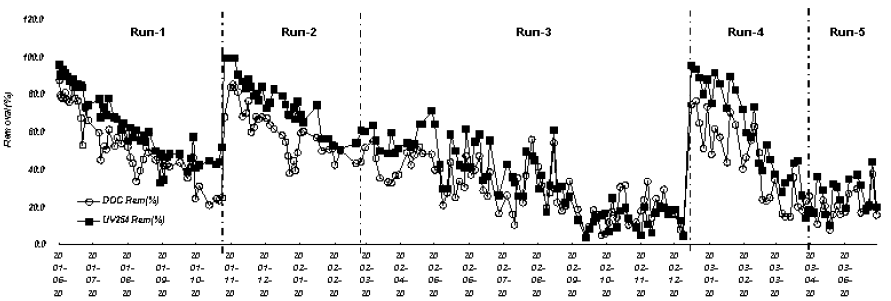

Figure 6 Variations of removal efficiency of DOC and UV_{254} by HCPAC-MBR

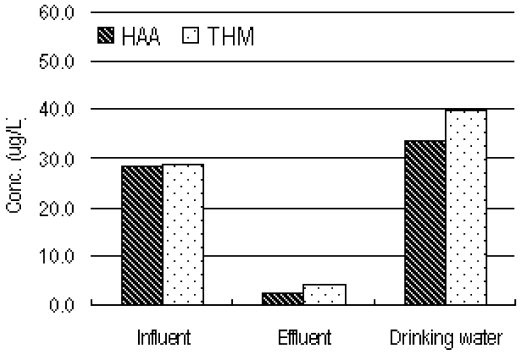

Figure 7 Removal of DBPs by HCPAC-MBR

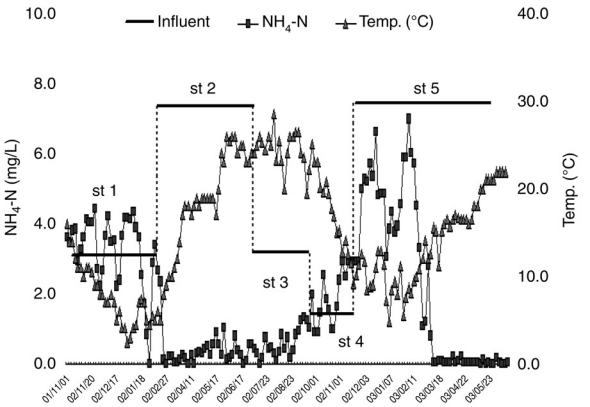

Figure 8 Removal of NH_3-N by HCPAC-MBR

NH_3-N removal. Figure 8 is the result of ammonia oxidation by the hybrid system. Ammonia concentration of the influent was controlled in the range 1.0 to 7.0 mg/L. Proper oxidation of ammonia was not observed during the first stage because of insufficient acclimation of ammonia oxidizing bacteria at the low water temperature. In the 2nd stage the almost complete oxidation was observed at increased water temperature. However, the ammonia oxidation deteriorated again in the 3rd stage. During this period the pH value was below 6.0 (5.0–5.8) and then the pH was adjusted at around 7.0 using sodium bicarbonate. Although stable ammonia oxidation could not be maintained by such operational troubles, it was observed that rapid reduction of ammonia concentration occurred even at water temperatures below 10 degrees Celsius in stage 5. Seo *et al*. (2002) have also reported excellent ammonia removal in their laboratory scale experiment of the HCPAC-MBR process. Therefore the HCPAC-MBR system has a large potential to be used for ammonia removal during the winter season when the concentration is high in the water supply source of the downstream Nackdong River basin.

Conclusions

The following conclusions were obtained from the long term operation of the pilot scale HCPAC-MBR process.

1. Among three types of PAC (lignite, charcoal, coconut shell), the coconut shell type PAC had better adsorption capability for DOC, HAA and THM. PAC concentration of 40 g/L and intermittent suction/idle time, 12 min/3 min, were the optimum operational parameters for the system.
2. At the flux of 0.36 m/d, the interval for replacement of the membrane was 2–3 months

with intermittent suction (12 min/3 min, for suction/idling). Air-backwashing once a day was very efficient in maintaining stable filtration for an extended period.

3. The organic removal by the HCPAC-MBR was 35–75% and 60–90% in terms of DOC and UV_{254}, respectively. It was evident that there was some combination between adsorption and biodegradation from the continuous removal efficiency of UV_{254} being higher than DOC through the experimental period.
4. Significantly high removal of HAA_5 (92.1%) and total THM (86.1%) allows the system to meet the stringent drinking water quality standard for DBPs. Although stable ammonia oxidation was not obtained, the HCPAC-MBR system showed a large potential for use in ammonia removal during the winter season.

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