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Effect of internal recycle rate on the high-strength nitrogen wastewater treatment in the combined UBF/MBR system

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Abstract An anaerobic/aerobic system combining an anaerobic upflow-sludge bed filter (UBF) and an aerobic membrane bioreactor (MBR) was operated to enhance organic and nitrogen removal efficiency. The internal recycle rate, which is one of the most important operation factors that affects overall removal efficiency, was varied from 100% to 300% of the influent flow. Under these conditions, the overall removal efficiencies of organic and nitrogen and characteristics of membrane fouling in the combined system treating the synthetic wastewater including high concentration of organics and nutrients were studied. As a result, nitrogen removal efficiency was increased to 67% when the internal recycle rate was 300% of influent flow rate. As the internal recycle ratio increased from 100% to 200%, protein content decreased by 17% and carbohydrate content increased by 12%. However, there was no remarkable difference in total extracellular polymeric substances (EPS) content. At the high recycle rate of 300%, the surface charge of sludge was decreased while hydrophobicity (specific ultraviolet absorbance, SUVA) was increased. The differences in SUVA and surface charge were 11% and 1%, respectively. It is concluded that SUVA and EPS composition were important parameters affecting membrane fouling in the combined system.
Keywords UBF; MBR; internal recycle rate; membrane fouling; hydrophobicity

Introduction

The wastewater of high-strength nitrogen with high organic matter content has been treated in anaerobic biological reactors due to the economic and other advantages such as the methane produced that can be used as energy. For these advantages, the anaerobic digestion technology has been widely applied for organic and solid removal (Rittmann and McCarty, 2001). Also as the water quality standards regarding nitrogen in effluent become more stringent, the effective removal of nitrogen to fulfill the discharge limits by nitrification and denitrification is necessary.

The technologies usually applied for nutrient removals are based on a spatial or temporal sequence of different anaerobic and anoxic, aerobic steps (Lacalle *et al.*, 2001). Recently, a direct integration of the denitrification and the anaerobic function in a single reactor has been proposed as an alternative complex system for simultaneous carbon and nitrogen removal (Akunna *et al.*, 1995; Hanaki and Polprasert, 1989; Kuroda *et al.*, 1988; Shin *et al.*, 1999). In addition, various configurations of anaerobic and aerobic reactors were applied for the treatment of high concentration of nitrogen wastewater (Barber and Stuckey, 2000; Bernet *et al.*, 2000). For the effective operation of the integrated treatment system, the internal recycle of oxidized nitrogen from the anaerobic reactor to the following aerobic reactor was essential and crucial factor affecting the overall nitrogen removal efficiency. In the previous study, the combined UBF/MBR system has been proved for the effective treatment of high-strength nitrogen wastewater. At the internal recycle ratio of Q (the influent flow rate), average removal efficiencies of organic and total nitrogen were 99% and 46%, respectively (Shin *et al.*, 2003).

For better performance, removal efficiencies of organic and nitrogen in the combined system was evaluated at the various internal recycle rates. Also various parameters affecting membrane fouling, which is one of the major obstacles in MBR coupled process (Lee *et al.*, 2003; Mukai *et al.*, 2000), was monitored.

Material and methods

Characteristics of the combined UBF/MBR system

As shown in Figure 1, the anaerobic UBF reactor having 6.27 L working volume (WV) was filled with ceramic filter media. Porosity and bulk density of the media were 75–80% and 0.03–0.37 g/cm^3, respectively. Influent was supplied to the bottom of the UBF reactor with the recycled MBR effluent containing oxidized nitrogen. The internal recycle ratio was adjusted to 100% of influent flow rate. The UBF reactor was operated at 35 ± 1 °C and the produced gas was collected in a gas collector. The seed granular sludge was taken from a brewery wastewater treatment plant.

In the aerobic MBR having 6.5 L WV, a hollow fiber membrane module was placed. The microfiltration membrane was made of polypropylene having a nominal pore size of 0.4 μm and an effective filtration area of 0.07 m^2. Initially, the bioreactor was filled with sludge from municipal wastewater treatment plant. For the easy build-up of nitrifying bacteria in the bioreactor, no sludge was discharged during the experiment period. Both reactors were connected in series with a recycling line from the MBR to the influent line of the UBF.

Characteristic of influent

As shown in Table 1, the UBF reactor was fed with the synthetic wastewater containing glucose and ammonia as the principal organic carbon and nitrogen sources respectively to study the effect of internal recycling from the UBF on removal efficiencies of organic and nitrogen. Influent COD and nitrogen concentrations were fixed at 14,500 mg/L (7.2 kg COD/m^3/d) and 1,000 mg/L (0.5 kg NH_4-N/m^3/d), respectively.

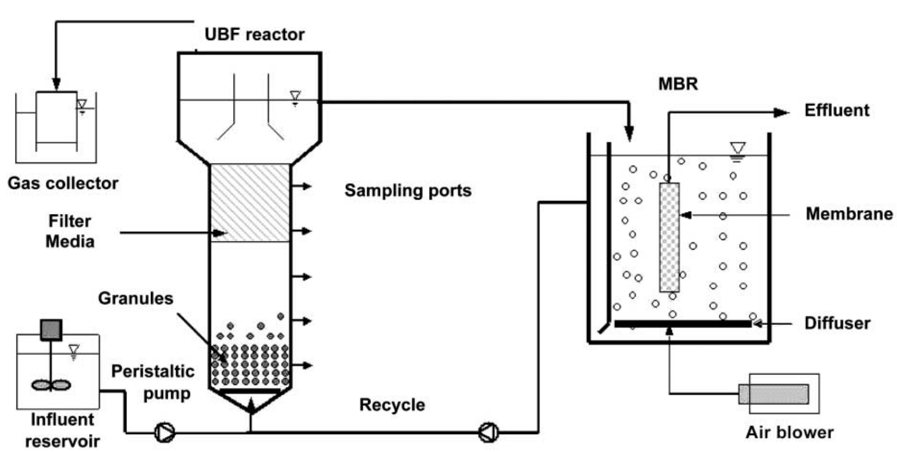

Figure 1 Schematic diagram of the combined UBF/MBR system

Table 1 Characteristics of the synthetic wastewater

Compound	Chemical formula	Molecular weight (g/mol)	Concentration (mg/L)
Glucose	$C_6H_{12}O_6$	180.0	14,500
Ammonium chloride	NH_4Cl	53.5	1,000
Calcium chloride	$CaCl_2.2H_2O$	147.0	0.368
Magnesium sulfate	$MgSO_4.7H_2O$	246.5	5.07
Manganese chloride	$MnCl_2.4H_2O$	197.9	0.275
Zinc sulfate	$ZnSO_4.7H_2O$	287.5	0.44
Ferric chloride anhydrous	$FeCl_3$	162.2	1.45
Cupric sulfate	$CuSO_4.5H_2O$	249.7	0.391
Cobalt chloride	$CoCl_2.6H_2O$	237.9	0.42
Sodium molybdate dehydrate	$Na_2MoO_4.2H_2O$	242.0	1.26
Yeast extract	–	–	30

Experimental conditions

As shown in Table 2, internal recycle ratio was varied from 100% to 300% of influent flow rate. Both of anaerobic and aerobic reactors were operated at the same hydraulic retention time (HRT) of 24 hours, except for phase 2.

Analytical methods

Suspended solids (SS), COD, TKN and NH_4-N concentrations were measured according to *Standard Methods* (APHA, 1998). Concentrations of various ions in solution were analyzed using ion chromatography (DX-120, DIONEX, USA). Total organic and inorganic carbons were determined by a TOC analyzer (DC-180, Dorhmann, Germany). The surface charge of microbial floc was determined by titration method. Polybrene and polyvinyl sulphate (PVSK) were used as the cationic and anionic standards, respectively, in the titration method. A known volume of sludge sample was diluted with ultra-pure water and mixed with an excess amount of 0.001 N polybrene standard solutions. Standard solution of 0.001 N PVSK was used to titrate against the excess amount of polybrene using a few drops of toluidine blue as an indicator; a subtle color change from blue to purple. An equal volume of polybrene diluted with the same amount of deionized distilled water was used as a blank. Then, the surface charge can then be determined from the following equation:

$$\text{Surface charge (meq/gVSS)} = \frac{(A - B) \times N \times 1000}{V \times M}$$

where, A is mol of PVSK added to the sample, N normality of PVSK, B ml of PVS added to blank, V ml of sample used, and M g VSS/L.

SUVA was used to estimate the hydrophobicity of supernatant. SUVA is the ratio of UV absorbance at 254 nm to dissolved organic carbon concentration. High SUVA indicates high degree of hydrophobicity due to its high aromaticity. The procedure of surface charge measurement was the same as described previously (Lee *et al.*, 2003). EPS was extracted from microbial floc using heat treatment (Morgan *et al.*, 1990). The extracted

Table 2 Experimental conditions

Phase	Recycle rate	HRT		Operation period
		UBF	MBR	
1	100%	24 hr	24 hr	1–15 days
2	200%	16 hr	16 hr	16–30 days
3	200%	24 hr	24 hr	31–41 days
4	300%	24 hr	24 hr	42–55 days

solution was analyzed for total carbohydrate and proteins. The sum of the amounts of total carbohydrates and proteins represented the total amount of EPS, which are the dominant components typically found in extracted EPS (Bura *et al.*, 1998; Frølund *et al.*, 1996). Carbohydrates and proteins in EPS were determined according to the phenol-sulfuric acid method with glucose as standard (Dubois *et al.*, 1956) and Folin method with bovine serum albumin as standard (Lowry *et al.*, 1951), respectively.

Results and discussion

Performance of the combined UBF/MBR system

Table 3 and Figure 2 present the process performance at various internal recycle rates. During the experimental period, higher organic and nitrogen removal was possible. When the internal recycle ratio was 100% (phase 1), average removal efficiencies of organic and total nitrogen were 99% and 47%, respectively.

In phase 2, ammonia nitrogen concentration of MBR effluent increased to 345 mg/L. Deterioration of effluent quality, in spite of the increased internal recycle ratio from 100% to 200%, was observed resulting from the decrease of HRT from 24 to 16 hours in both reactors. In phase 3, the internal recycle ratio was adjusted to 200% and HRT was increased from 16 hours to 24 hours resulting in the improvement of total nitrogen removal efficiency (from 41% to 50%). Finally, total nitrogen removal efficiency was increased to 67% when the internal recycle rate was 300% of influent flow rate in phase 4.

Membrane fouling at various internal recycle rates

There were various biological, physical and chemical factors affecting membrane fouling in the activated sludge. In the previous study, which compared the membrane fouling characteristics between combined and unit process with the internal recycle ratio of Q, it was found that the total amount of EPS, hydrophobicity of supernatant and surface charge of sludge were sensitive factors while there was no significant change in particle size and colloid distribution of the sludge and supernatant, respectively (Shin *et al.*, 2003). EPS matrix is heterogeneous, in which a variety of polymeric materials have been found; such as carbohydrates, proteins, lipids and nucleic acids. In this study, however, the sum of total carbohydrates and proteins was considered to represent the total amount of EPS because these were the dominant components typically found in extracted EPS (Bura *et al.*, 1998; Frølund *et al.*, 1996). Figure 3 shows the concentrations of carbohydrates and protein in microbial floc. The total amount of EPS on microbial floc were 65.1, 64.5 and 68.8 mg/g VSS at various internal recycle ratio of Q, 2Q and 3Q, respectively.

Even though there was only small variation in total amount of EPS content, as the internal recycle rate increased from 100% to 200%, protein content decreased by 17% and carbohydrate content increased by 12%. With a high food to microorganism (F/M) ratio in aerobic reactor due to the higher COD concentration of UBF effluent, EPS carbohydrate content in sludge increased, which reflected the available carbon.

Table 3 COD and T-N removal efficiencies under various operation conditions

	Item	Phase 1	Phase 2	Phase 3	Phase 4
Effluent (mg/L)	COD	100.9	178.3	158.9	98.5
	NH_4-N + NO_3-N	533.3	597.9	507.3	332.1
Removal (%)	COD	99	99	99	99
	T-N	47	41	50	67

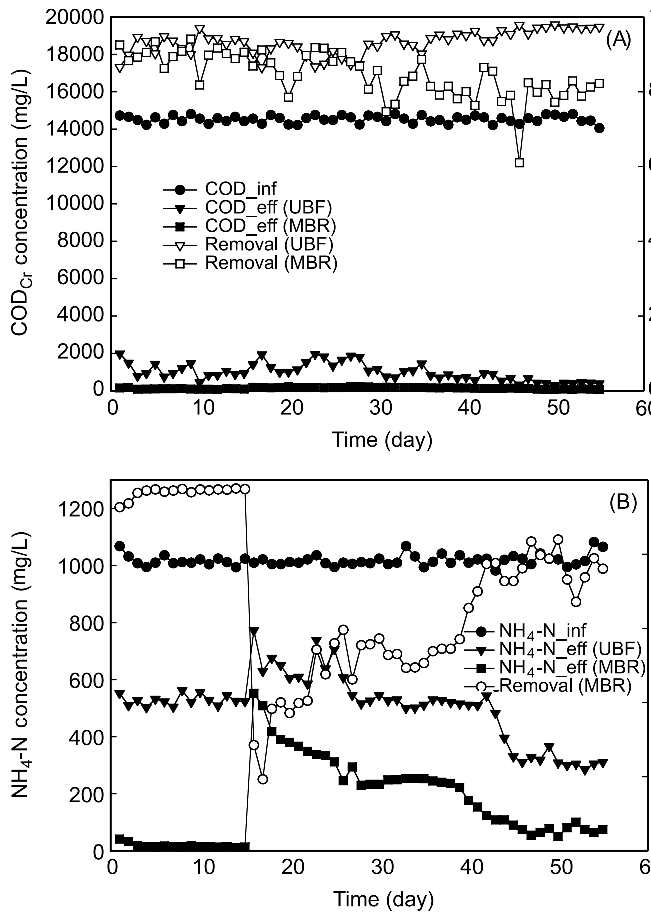

Figure 2 COD (A) and nitrogen (B) removal in the anaerobic/aerobic combined system

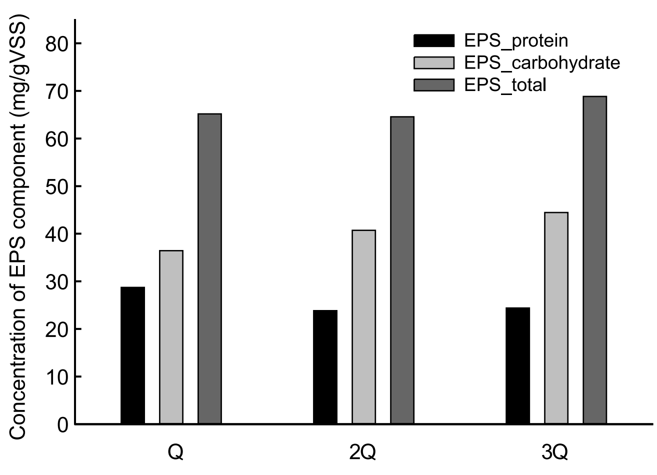

Figure 3 Concentrations of EPS components at various internal recycle rates

Table 4 Surface properties of microbial floc and supernatant in various phases

Phase	Internal recycle rate	Day	SUVA ($m^{-1}\,mg^{-1}\,L$)	Surface charge (meq./g VSS)
1	100%	15	2.81	−0.3211
3	200%	41	2.94	−0.3182
4	300%	55	3.12	−0.3179

As shown in Table 4, hydrophobicity and surface charge were measured to evaluate the surface properties of supernatant and microbial floc, respectively. As a result, the surface charge of sludge and hydrophobicity increased as the internal recycle rate increased from 100% to 300%. The differences of SUVA and surface charge between phase 1 and phase 3 were 11% ($0.31\,m^{-1}\,mg^{-1}\,L$) and 1% (0.0032 meq./g VSS), respectively. It was concluded that EPS composition and SUVA was more sensitive than hydrophobicity or total EPS content, which was also the most sensitive parameter in the previous study (Shin *et al.*, 2003).

Conclusions

The effect of internal recycle ratio on the combined anaerobic/aerobic system, which is fed with high-strength nitrogen wastewater, was studied at various recycle rates of 100%, 200% and 300% of influent flow rate. As it was increased to 300%, total nitrogen removal was increased to 67%. At the relatively short HRT of 24 hours, quite stable organic and nitrogen removal was possible in both reactors compared to previous researches. As the internal recycle rate increased, protein content decreased by 17% while carbohydrate content increased by 12%. Also, at a high recycle rate of 300%, the surface charge of sludge and hydrophobicity were increased.

Total EPS content and hydrophobicity were important parameters in the previous study. Membrane fouling was more severe in the combined process MBR than unit process one with the internal recycle rate of 1Q. However, in this study, it was concluded that EPS composition and SUVA was a more sensitive parameter than surface charge and total EPS content, with respect to the change of recycle rate 100% to 300%. Also it was recommended to operate with enough HRT (in this study longer than 24 hours), which could lead to effective organic and nitrogen removal, to minimize the membrane fouling in the combined UBF/MBR process.

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