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Publication date:
2011

Document Version
Publisher's PDF, also known as Version of record

Simultaneous removals of azo dye and nitrogenous compounds by a membrane-aerated biofilm

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Keywords: azo dye; membrane-aerated biofilm; biological nitrogen removal

Mineralization of azo dyes, main constituents of textile dye wastewaters, necessitates sequential anaerobic and aerobic biological reactions. The first reaction is the reductive cleavage of azo-bonds (-N=N-) whereas the second reaction is oxidation of the cleaved intermediates, less biodegradable and more toxic than the original azo dyes. Therefore, conventional activated sludge systems are not appropriate because the accumulation of the intermediates can inhibit microbial activity. In addition, textile dye wastewater contains not only ammonium released via azo dye mineralization steps but also high concentration of nitrogenous compounds as an auxiliary agent, which requires biological nitrogen removal. Taken together, the use of redox stratification in biofilms, where both anaerobic and aerobic regions are present with biofilm depth, is of promise to treat azo dye and nitrogenous compounds in a single reactor. Nevertheless, control of the redox stratification is still a challenge and lack of control potentially deteriorates removal performance of azo dye compounds.

The use of biofilm grown on a gas-permeable membrane, *i.e.*, membrane-aerated biofilm (MAB), can be promising for simultaneous removals of azo dye and nitrogenous compounds. The MAB has a configuration where oxygen is permeated from the gas phase to the biofilm base without bubble formation whereas azo dye and nitrogenous compounds diffuse from the surface of the biofilm. By adjusting gas pressure or flow rate in the gas phase, oxygen loading to the biofilm can be controlled. Such counter-current loadings of oxygen and contaminants are conducive to separate manipulation, permitting intentional control of redox stratification in a biofilm (Terada et al. 2007). Therefore, the objectives of this study were to investigate the feasibility of simultaneous removals of azo dye and nitrogen with an MAB, to monitor the microbial community and to elucidate the spatial distribution of dominant bacteria in the biofilm.

A membrane-aerated biofilm reactor (MABR) with a volume of 0.63 L was constructed. A membrane module consisting of 960 composite hollow-fiber membranes with each inner/outer fiber diameter of 200/280 μm (Model MHF 3504, Mitsubishi Rayon Co. Ltd., Tokyo) was mounted in the MABR and air was supplied from the membrane lumen. The synthetic wastewater, comprised of acid orange 7 (AO7) of 50 mg-C/L as a representative azo dye, ammonium of 77 mg-N/L as an auxiliary agent for textile dyes and sodium acetate of 240 mg-C/L as an external electron donor, was continuously fed to the MABR. Complete mixing condition was ensured at a wastewater flow rate of 0.1 L/min with a recirculation pump. The wastewater loading rates and supplied air pressures are shown in Table 1. Concentrations of AO7, the intermediates, *i.e.*, sulfanilic acid (SA) and 1-amino-2-naphthol (AN), and nitrogenous compounds were monitored for 180 days. The MABR operation was initiated with sequential inoculation of three biomasses enriched by AO7, SA and AN, respectively. Transition and spatial distribution of microbial community in the biofilm was confirmed by denaturing gradient gel electrophoresis (DGGE) and fluorescence *in situ* hybridization (FISH).

Removal efficiency of AO7 was converged to approximately 70% in Period I after 70 day while SA and AN were not detected (Figure 1A). This indicates that AO7 reduction and SA/AN oxidation concomitantly occurred in the biofilm. Nitrogenous compounds were also removed in the reactor (Figure 1B). Total nitrogen (T-N) removal efficiency reached approximately 95% in Period I, suggesting occurrence of simultaneous nitrification and denitrification. In Period II, AO7 removal efficiency increased up to more than 95%, which has been stable until the end of the experiment. In Period III, NH_4^+-N concentration in effluent increased probably due to shortage of oxygen supply and excessive biofilm growth. Increase in air pressure successfully reduced NH_4^+-N concentration in effluent (Figure 1B), improving T-N removal efficiency from 75% (Period IV) to 90% (Period VII). This indicates that adjustment of air pressure in the MABR allowed improving nitrogen removal without impairing AO7 removal. The increase in air pressure slightly improved AN but not SA removal, suggesting that SA oxidation is one of the limiting steps for AO7 mineralization. In sum, removal efficiencies of AO7, SA, AN and T-N were 95%, 80%, 100% and 80% at carbon and nitrogen volumetric loading rates of 510 g-C/m^3/day and 118 g-N/m^3/day, respectively.

DGGE and subsequent gel excision of intensified bands revealed that predominant bacteria in the inocula for AO7, SA and AN removals were affiliated to *Acinetobacter* spp., uncultured beta proteobacteria and *Thermomonas* spp., respectively. *Acinetobacter* spp., putative AO7 degraders, have been predominant in the biofilm but the others were mainly replaced with *Zoogloea* spp, *Rhizobium* spp. and *Nitrosospira* spp., respectively. This infers that these enriched bacteria are potential SA/AN degrading and/or ammonia-oxidizing bacteria in the biofilm. FISH observations exhibited ammonia-oxidizing bacteria were predominant adjacent to a membrane surface where oxygen is supplied, supporting NH_4^+-N removal in the biofilm. Spatial distribution of the predominant bacteria identified by the DGGE will be addressed in the presentation.

Table 1 MABR experimental conditions for simultaneous removals of azo dye and nitrogen.

Period	HRT [day]	Air pressure [kPa]	AO7 loading rate [g-C/m^3/day]	Organic-C loading rate [g-C/m^3/day]	Nitrogen loading rate [g-N/m^3/day]
I	0.97	0.75	59	306	71
II	0.80	0.75	72	374	86
III	0.58	0.75	99	510	118
IV	0.58	0.9	99	510	118
V	0.58	1.1	99	510	118
VI	0.58	3.0	99	510	118
VII	0.58	8.0	99	510	118

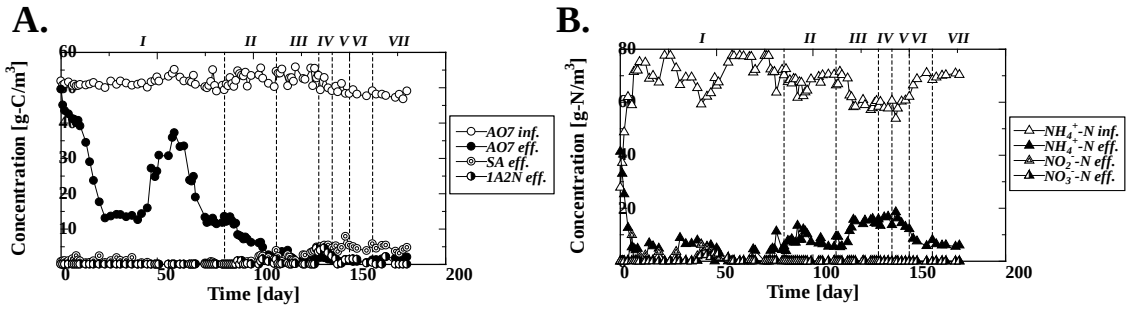

Figure 1 Time courses of influent and effluent concentrations of azo dye and the intermediates (A) and nitrogenous compounds (B).

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