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NITROGEN REMOVAL FROM WASTEWATERS BY A BIO-REACTOR WITH PARTIALLY AND FULLY SUBMERGED ROTATING BIOFILMS

Y. Watanabe*, D. Y. Bang**, K. Itoh*** and K. Matsui†

* *Department of Civil and Environmental Engineering, Miyazaki University, Miyazaki 889-21, Japan*
** *Department of Civil Engineering, Tohoku University, Aobaku, Sendai 980, Japan*
*** *Toshiba Co. Ltd., Fuchyu, Tokyo 183, Japan*
† *Nishihara Environmental Sanitation Research Co. Ltd., Minatoku, Shibaura, Tokyo 108, Japan*

ABSTRACT

This paper concerns simultaneous nitrification and denitrification in a completely mixed bio-reactor with partially and fully submerged rotating biological contactors. The bio-reactor is designed to cause the nitrification and denitrification in partially and fully submerged biofilms, respectively. An experimental investigation was made into the effect of organic material and ratio of influent organic carbon to ammonia nitrogen concentrations(C/N ratio) on the efficiency of simultaneous nitrification and denitrification in the bio-reactor. Settled municipal wastewater and synthetic wastewater containing ammonia nitrogen and organic material such as acetate, ethylene-glycol,phenol and poly-vinyl-alcohol(PVA) were fed into the experimental units. A biofilm dominated by nitrifiers developed on the partially submerged contactors, while a biofilm dominated by heterotrophs developed on the fully submerged contactors. A micro-aerobic environment was formed and biological denitrification occurred in the submerged biofilm. In the municipal wastewater treatment where the influent C/N ratio was around 3.5, the maximum nitrogen removal efficiency was about 60 %. Acetate and ethlene-glycol were effectively used as the organic source of the denitrification. The ability to aerobically degrade PVA was induced by phenol. Once the bacteria inhibiting the biofilm gained the ability to degrade PVA, PVA became an effective organic source of the denitrification.

KEYWORDS

Rotating biological contactor, partially and fully submerged biofilms, organic compounds, nitrification, denitrification.

INTRODUCTION

Biological nitrogen removal has usually been carried out by a multi-staged system consisting of the reactors for nitrification and denitrification. Masuda, Watanabe and Ishiguro(1987,1991) reported that nitrifiers and denitrifiers coexist throughout the whole depth of the biofilm attached to a bio-reactor treating wastewater containing organics and ammonia nitrogen. They (1979,1982,1990,1992) studied the phenomenon of simultaneous nitrification and denitrification occurring in a micro-aerobic biofilm, which was attached to a partially submerged rotating biological contactor (RBC) with a reduced partial pressure of oxygen in the air phase. This research was concerned with a bio-reactor in which simultaneous nitrification and denitrification occur in a micro-aerobic biofilm. The authors(Watanabe,Itoh and Matsui,1993) have also developed a bio-reactor with partially and fully submerged rotating biofilms. This bio-reactor was designed to produce nitrification and denitrification mainly in the partially and fully submerged biofilms, respectively.

This paper reports on the performance of the bio-reactor in treating settled municipal wastewater and synthetic wastewater with acetate,ethylene-glycol, phenol and poly-vinyl-alcohol as the organic source. A kinetic analysis of the data is also described.

MATERIALS AND METHODS

Fig.1 shows the schematic description of a bio-reactor with partially and fully submerged rotating contactors. Based on previous investigations (Watanabe et al.,1990), the thickness of the liquid boundary layer,which offers mass transfer resistance to the biofilm, is much larger in the fully submerged biofilm than in the partially submerged biofilm. For example, it is estimated to be about 100μm for the partially submerged biofilm and about 250μm for the fully submerged biofilm, respectively, at a rotating speed of 2 rpm. Therefore, a micro-aerobic environment is formed within the submerged biofilm even though the dissolved oxygen concentration is 3 to 4 mg/L. Two bench scale units were used. Their dimensions are described in Table 1. Unit 1 was installed in a municipal wastewater treatment plant to treat settled municipal wastewater without temperature control. The C/N ratio of the settled municipal wastewater was around 3.5. It was pretreated by a partially submerged RBC, when the effect of the influent C/N ratio on the nitrogen removal efficiency was examined. Unit 2 was installed in a constant-temperature room to treat synthetic wastewater. Four kinds of synthetic wastewater were used. They contained ammonia nitrogen, organic material, and trace elements. Acetate, ethylene-glycol,phenol and PVA were used as the organic material. Settled municipal wastewater was fed into the bio-reactor to grow the biofilm before the experiment with the synthetic wastewater was started.

At the end of the experiment with the settled municipal wastewater, a part of the biofilm was removed from the partially and fully submerged contactors to measure the activity of the nitrification and denitrification. The removed biofilm was destroyed by supersonic power(400μA,

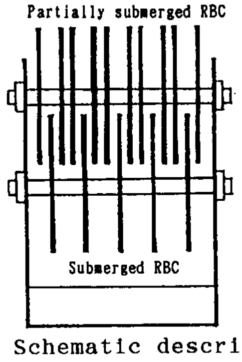

Fig.1 Schematic description of bioreactor

Table 1 Dimensions of units

Items	Unit 1	Unit 2
Effective volume(L.)	33	6.4
Contactor diameter(cm)	30	16
Contactor number		
partially submerged	8	10
fully submerged	4	5
Rotating speed(rpm)	2	7-14

30 seconds), then added to an aerated jar which contained ammonia nitrogen ,inorganic carbon and trace elements for measuring the nitrifying activity. The destroyed biofilm was added to a stirred jar which contained nitrate nitrogen, the acetate and trace elements for measuring the denitrifying activity.

RESULTS AND DISCUSSION

(1) Reactor characteristics

Run 1 was conducted using Unit 2 to examine the basic performance of the bio-reactor. The synthetic wastewater containing 25 mg/L of NH_4-N, 70 mg/L of CH_3COOH as TOC and the trace elements. The water temperature was kept at 25°C and the hydraulic retention time (HRT) was varied from 3 to 7 hours. Fig.2 shows the efficiencies of the nitrifi-

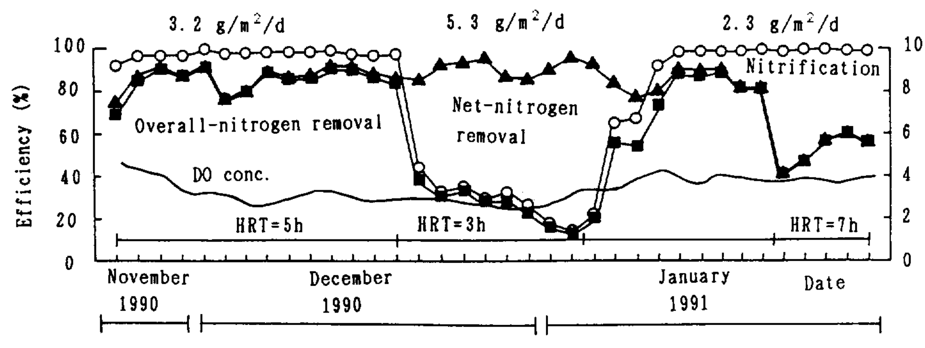

Fig.2 Performance of bio-reactor

cation, net nitrogen removal and overall nitrogen removal. Fig.2 clearly indicates the following characteristics of the bio-reactor: (1) At an HRT of 5 hours,corresponding to a TOC loading to the partially submerged biofilm of 3.2 g/m²/d, very high nitrogen removal efficiency was obtained,(2) At a higher organic loading, nitrogen removal was limited by nitrification, while nitrogen removal was limited by denitrification at a lower organic loading, and (3) In spite of a rather high dissolved oxygen concentration, the denitrification was quite active.

In Run 2, settled municipal wastewaster was fed into Unit 2. No control of the water temperature was attempted. The average water temperature was about 17 °C during the experiment. The HRT was changed to achieve a nitrification efficiency of 80 % at a given influent C/N ratio. Fig.3 shows the effect of the influent C/N ratio and HRT on nitrogen removal efficiency. At an influent C/N ratio of around 3.5, the overall nitrogen removal efficiency was about 60 % when the HRT was

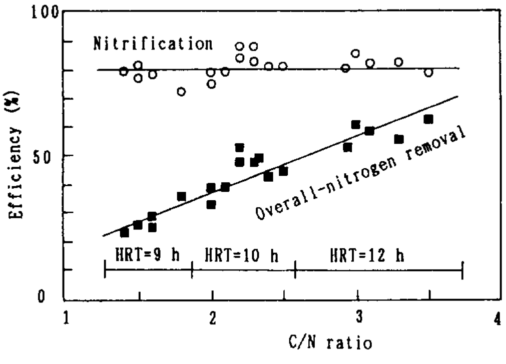

Fig.3 Nitrogen removal efficiency of municipal wastewater

set at 12 hours,corresponding to a TOC and NH_4-N loading to the partially submerged biofilms of 2.3 and 1.1 g/m^2/d,respectively. Table 2 shows the specific activity of the biofilms measured at the end of Run 2.

(2) Degradation of acetate and ethylene-glycol

In Run 3, the diluted supernatant of a night soil digester was used as the influent to Unit 2. Its NH_4-N and TOC concentrations were approximately 120 mg/L and 50 mg/L, respectively. In order to increase the influent C/N ratio, acetate or ethylene-glycol was added to the influent. The water temperature and HRT were kept at 25 °C and 24 hours,respectively. Figs.4 and 5 show the effect of the influent C/N ratio on the efficiency of nitrification and nitrogen removal. The optimum C/N ratio giving maximum nitrogen removal efficiency exists.The optimum C/N ratio of the easily biodegradable acetate was larger than that of the less-easily biodegradable ethylene-glycol. A batch experiment was carried out during the experimental run using acetate as the organic source (see Fig.6). As the oxidized nitrogen produced in the partially submerged biofilm was reduced to gaseous nitrogen, such as N_2, in the fully submerged biofilm, no accumulation of NO_2-N and NO_3-N occurred. When the acetate and ethylene-glycol were used as the organic source, the stoichiometry of the denitrification reaction was described by Eqs.1 and 2,ignoring cell synthesis. From Eqs.1 and 2, it was calculated that 1.14 g and 0.86 g of organic carbon are needed to reduce 1 g of NO_3-N. Fig.7 shows the re-

Table 2 Biofilm activities

Biofilms	Nitrification	Denitrification
Partially submerged biofilm	1.5×10^{-2} 1/h	0
Fully submerged biofilm	2.2×10^{-3} 1/h	2.7×10^{-2} 1/h

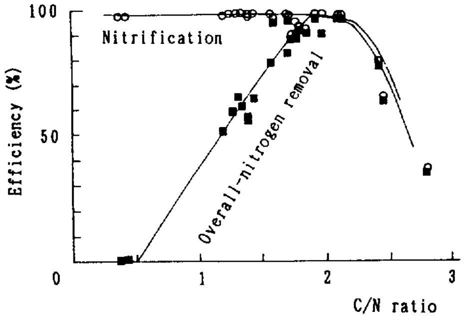

Fig.4 Effect of C/N ratio with ethylene-glycol

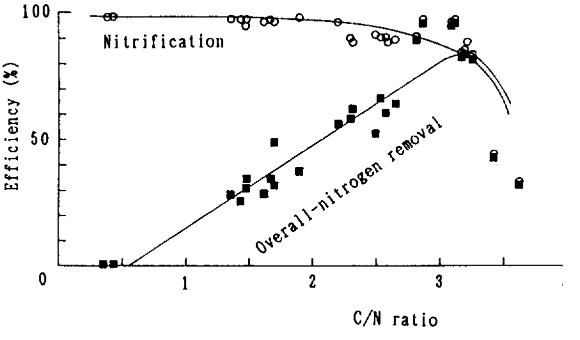

Fig.5 Effect of C/N ratio with acetate

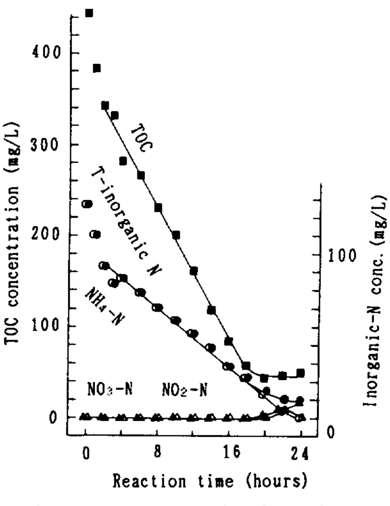

Fig.6 Result in batch experiment

$$6NO_3^- + 4CH_3COOH \rightarrow 3N_2 + 8CO_2 + 6H_2O + 4OH^- \quad (1)$$

$$2NO_3^- + HOCH_2CH_2OH \rightarrow N_2 + 2CO_2 + 2H_2O + 2OH^- \quad (2)$$

lationship between the removed TOC and the total nitrogen concentrations when acetate was used as the organic source of denitrification. Fig.8 shows a similar relationship when ethylene-glycol was used as the organic source of denitrification. The stoichiometry line was produced as explained above. These experimental results demonstrated that almost the same amount of ethylene-glycol was aerobically and anoxically degraded, however, more acetate was aerobically degraded than anoxically degraded.

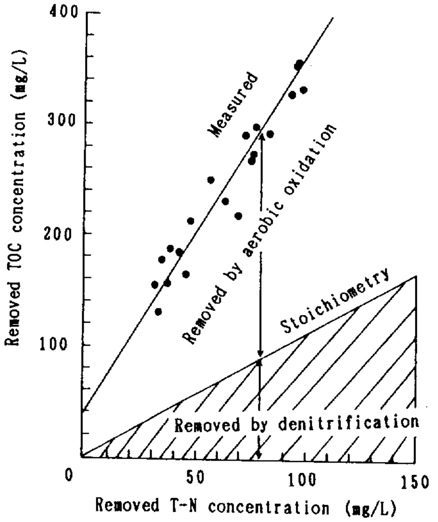

Fig.7 Removed TOC vs. removed T-N (Organic source=acetate)

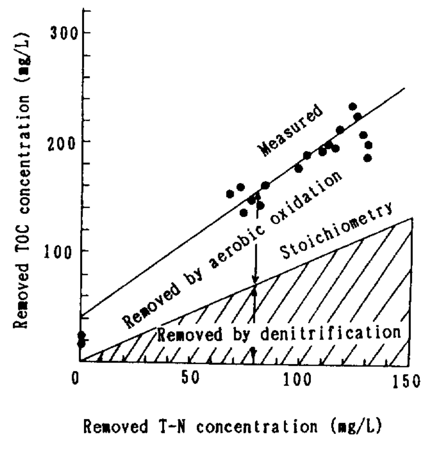

Fig.8 Removed TOC vs. removed T-N (Organic source=ethylene-glycol)

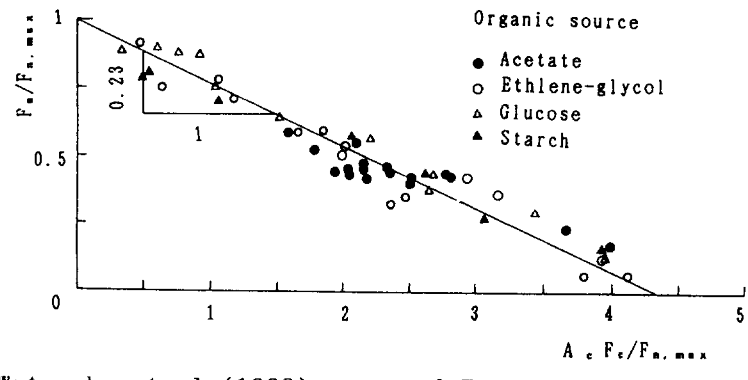

Fig.9 Plots of data followed by Eq.3

Watanabe,et al.(1982) proposed Eq.3 to express the relationship between the nitrification and organic oxidation rates under oxygen limitation conditions.

$$\frac{F_n}{F_{n,max}} = 1 - \left(\frac{A_c}{A_n}\frac{F_c}{F_{n,max}}\right) = 1 - \left(\frac{1}{4.33}\right)\left(\frac{A_c F_c}{F_{n,max}}\right) \quad (3)$$

where Fn= ammonia removal rate(g/m^2/d), Fn,max= maximum ammonia removal rate(g/m^2/d), Fc= organic oxidation rate(g/m^2/d), Ac= oxygen requirement of organic oxidation(g/g), An= oxygen requirement of nitrification(g/g)

This equation was derived from the hypothesis that the amount of oxygen transferred to the biofilm under oxygen limitation is kept constant at a given operating condition.
Fig. 9 shows the plots of the experimental data obtained in Run 2 along with the previous data (Watanabe, et al.,1984).

(3) Degradation of phenol and PVA

In Run 3, Phenol and poly-vinyl-alcohol(PVA) or only PVA was fed into Unit 2. Before Run 3 was started, the degradability of phenol and PVA under anoxic or aerobic conditions was examined using a closed- type partially submerged RBC. The experiment under anoxic conditions was continued for 73 days. The degree of polymerization of PVA was 1500 and 100 g of PVA is equivalent to 49 g TOC. Fig.10 shows the experimental results. It demonstrates that phenol was easily degraded to

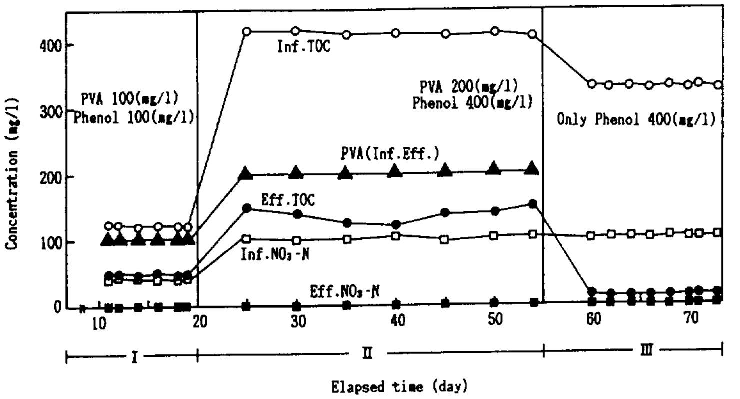

Fig.10 Effect of phenol and PVA as organic source in denitrification

reduce NO_3-N ,but PVA was not degraded under anoxic conditions. At an elapsed time of 74 days, the cover was removed to operate the RBC under aerobic conditions. The degrading ability of PVA increased with increased time, as seen in Fig.11. Nakamura and Miyaji(1992) reported that an enzyme responsible for trichloroethylene degradation was conjectured to be phenol hydroxylase. The result shown in Fig.11 may indicate that the degrading ability of PVA was induced by phenol. Fig.12 shows the experimental results of Run 4-1(influent phenol conc.=100 mg/L,influent PVA conc.=200 mg/L,influent NH_4-N conc.= 40 mg/L, influent C/N ratio=3.5) and Run 4-2 (influent phenol conc. and C/N ratio were increased to 200 mg/L and 5.2, respectively). It demon-

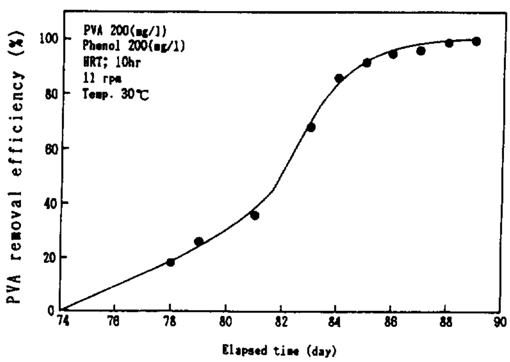

Fig.11 Increase in PVA degrading ability with elapsed time

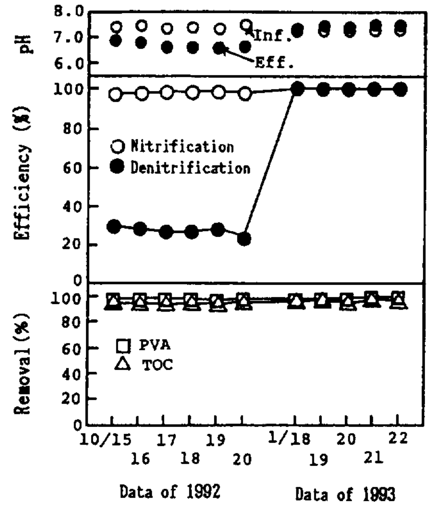

Fig.12 Reactor performance with phenol and PVA

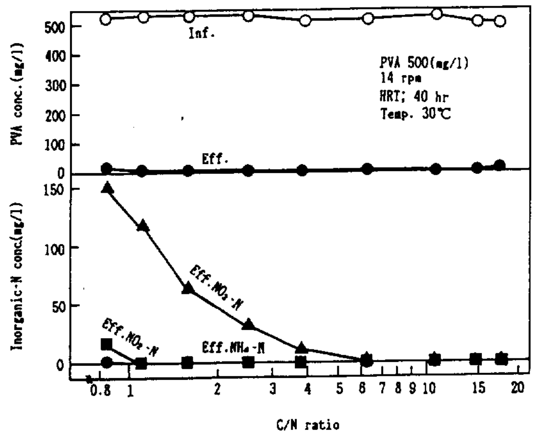

Fig.13 Effect of PVA on denitrification in varing C/N ratios

strated that most of the PVA was aerobically degraded and that phenol was used as the organic source of the denitrification in Run 4-2. In Run 4-3, only PVA was added to the influent at a concentration of 500 mg/L. The influent NH_4-N concentration was varied to adjust the influent C/N ratio. As shown in Fig.13, PVA was degraded mainly aerobically at the lower influent C/N ratio,while at the higher influent C/N ratio, it was aerobically and anoxically degraded. In Run 4-4, only PVA was added to the influent and the influent C/N ratio and water temperature were fixed at 3.0 and 20 °C,respectively. The HRT was gradually increased from 6 to 30 hours. Fig.14 shows the relationship between the efficiency of nitrification, overall nitrogen removal and PVA removal and the HRT. If the HRT is set at 30 hours, a high removal efficiency of nitrogen and PVA is achieved at a water temperature of 20 °C.

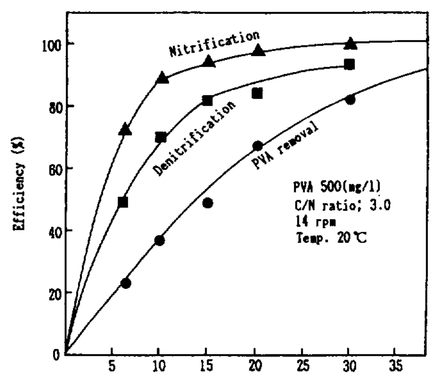

Fig.14 Effect of HRT on reactor performance

CONCLUSIONS

Two bench scale bio-reactors with partially and fully submerged rotating contactors were used to cause simultaneous nitrification and denitrification in a single unit. A biofilm dominated by the nitrifiers was developed on a partially submerged contactor. The biofilm was mainly inhibited by heterotrophs capable of denitrification which was attached to the fully submerged contactor. The experimental results demonstrated that the nitrogen removal efficiency in the bio-reactor was very high and the optimum influent C/N ratio existed to cause simultaneous nitrification and denitrification. The PVA,non-biodegradable organics, was degraded in the partially submerged biofilm when the biofilm bacteria were induced by the addition of phenol. Then, the degraded organics were used as the organic source of denitrification. A kinetic analysis of the data predicted that the amount of oxygen used for the aerobic oxidation of the organics and ammonia nitrogen under oxygen limiting conditions would be constant at a fixed operating condition.

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REFERENCES

Masuda,S.,Watanabe,Y. and Ishiguro,M.(1979):NITROGEN REMOVAL BY ROTATING BIOLOGICAL CONTACTORS(Part 1), J. of Japan Sewage Works Association, Vol.16,No.187,pp.24-32 (in Japanese)

Masuda,S.,Watanabe,Y. and Ishiguro,M.(1982):SIMULTANEOUS NITRIFICATION AND DENITRIFICATION IN A ROTATING BIOLOGICAL CONTACTOR, Proc. of the 1st International Conference on Fixed Biofilm Processes, pp.132-152

Masuda,S.,Watanabe,Y. and Ishiguro,M.(1987):PROPERTIES OF BIOFILM DEVELOPED IN ROTATING BIOLOGICAL CONTACTORS, J.of Japan Sewage Works Association,Vol.24, No.278, pp.19-31 (in Japanese)

Masuda,S.,Watanabe,Y. and Ishiguro,M.(1991):BIOFILM PROPERTIES AND SIMULTANEOUS NITRIFICATION AND DENITRIFICATION IN AEROBIC ROTATING BIOLOGICAL CONTACTORS, Water Science & Technology, Vol.23, No7-9 pp.1355-1363

Nakamura,K and Miyaji,Y.(1992):DEGRADATION OF TRICHLOROETHYLENE BY A GENETICALLY ENGINEERED MICROORGANISM CONTAINING A PHENOL HYDROXYLASE GENE, Proc. of Environmental Engineering Research, Vol.29, pp.17-27 (in Japanese)

Watanabe,Y.,Masuda,S.,Nisidome,K. and C. Wantawin(1984):MATHEMATICAL MODEL OF SIMULTANEOUS ORGANIC OXIDATION< NITRIFICATION AND DENITRIFICATION IN ROTATING BIOLOGICAL CONTACTORS, Water Science & Technology, Vol.17, No2/3, pp.385-397

Watanabe,Y.,Lee,C.,Koike,M. and Ishiguro,M.(1990):NITRIFICATION KINETICS AND SIMULTANEOUS REMOVAL OF BIOMASS AND PHOSPHOROUS IN ROTATING BIOLOGICAL CONTACTORS, Water Science & Technology, Vol.22, No.3/4, pp.169-178

Watanabe,Y.,Masuda,S. and Ishiguro,M.(1992):SIMULTANEOUS NITRIFICATION AND DENITRIFICATION IN MICRO-AEROBIC BIOFILMS, Water Science & Technology, Vol.26, No.3/4, pp.511-522