

In-situ Denitrification Process with Continuous Injection of Electrolytic Hydrogen and Oxygen

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1. INTRODUCTION

Nitrate contamination in groundwater has been detected in many places around the world (Follett, 1989). Its effect on human health has received considerable attention. In order to remediate the contaminated groundwater and protect it as a drinking water source, an *in-situ* denitrification process has been studied. In this study, a continuous experiment was conducted to evaluate the mass balances and removal performances of nitrate under different operating conditions.

2. MATERIALS AND METHODS

Fig. 1 shows an experimental apparatus used in this study. An artificial aquifer was designed for *in-situ* denitrification. The dimensions of the reactor were 2050 mm in length, 75 mm in width, and 860 mm in height, respectively. The aquifer was filled with glass beads ($\phi=2$ mm). There were 18 ports on one side as shown in Fig. 1. In experiment, an enriched denitrifying culture was injected into the ports 1 to 9 while extracts from soil/water mixture into the ports 10 to 18. After seeding, feed solution, hydrogen gas and oxygen gas were injected from left and at the bottom of port 6 and port 15, respectively. Experimental conditions (Runs 1~3) were shown in Table 1.

Mass balance of denitrification was taken based on the following equation:

$$Q_{H_2} = Q_{De} + Q_{O_2} + Q_{Ex} + Q_{Ex, gas} \quad (1)$$

Where Q_{H_2} is the amount of hydrogen gas injected per day, and Q_{De} and Q_{O_2} are equivalent amounts of hydrogen used for denitrification and oxygen utilization, and Q_{Ex} and $Q_{Ex, gas}$ are those discharged with bulk liquid and gas effluent, respectively.

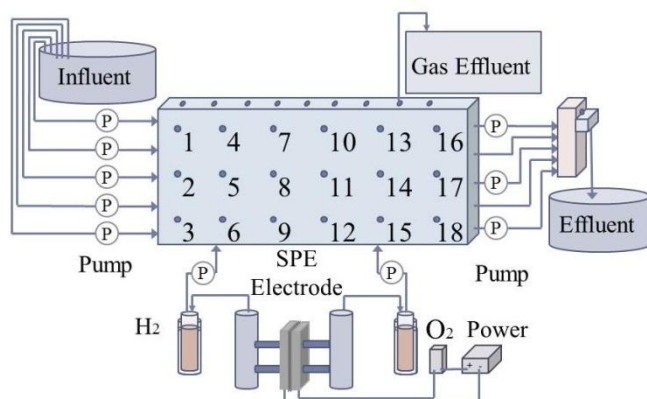


Fig. 1 Experimental apparatus

Table 1 Experimental conditions

	Run-1	Run-2	Run-3
Flow rate (Q)	5.8±0.1 l/d		
HRT	2.65~2.74 d		
Temperature	20±2 °C		
Buffer (NaHCO ₃)	0.85 mmol/l	8.50 mmol/l	
Nitrate	1.07±0.10 mmol/l		
H ₂ gas	8.6±1.0 mmol/d	15.8±1.9 mmol/d	
O ₂ gas	6.2±1.2 mmol/d		

3. RESULTS AND DISCUSSION

Fig. 2 shows time courses of nitrate and nitrite at positions of 0 m (Influent), 0.75 m, 1.05 m and 2 m (Effluent), respectively. In Run 1, nitrate concentration decreased and nitrite concentration increased near the H₂ injection zone (ports 6 and 9). However, in Run 2, the accumulation of nitrite shrunk and stable denitrification occurred. It was considered that an addition of carbonate buffer lowered pH (Fig. 3) and then enhanced denitrification rates of nitrite to nitrogen gas. Nitrite concentrations were less than 0.03 mmol/l in Runs 2 and 3. In Run 3, where the amount of injected H₂ was increased, nitrate concentration tended to decrease. In addition, nitrification of residual nitrite to nitrate occurred in place around the O₂ injection zone (port 15). Hydrogen mass balances by Eq. (1) satisfied around 100% in Runs 2 and 3 (Fig. 4).

Keywords: *In-situ*, Denitrification, Hydrogen, Oxygen

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TOC concentrations tended to increase in a small amount at the H₂ injection zone and then decrease to around 2.5 mg/l (in Runs 2 and 3) at the O₂ injection zone as well as in the effluent. Sufficient amount of DO was supplied in the O₂ injection zone (Fig. 5). These results indicated that denitrification and oxygenation/aerobic biological reaction zones were achieved in the present *in-situ* process.

Furthermore, similar trends of turbidity and chromaticity to TOC were observed, demonstrating they increased and then decreased to around 0.3 NTU and 2.5 PCU (in Runs 2 and 3) in the effluent, respectively. These results indicated the decomposition of suspended solid in oxygenation/aerobic oxidation zone might occur in the process.

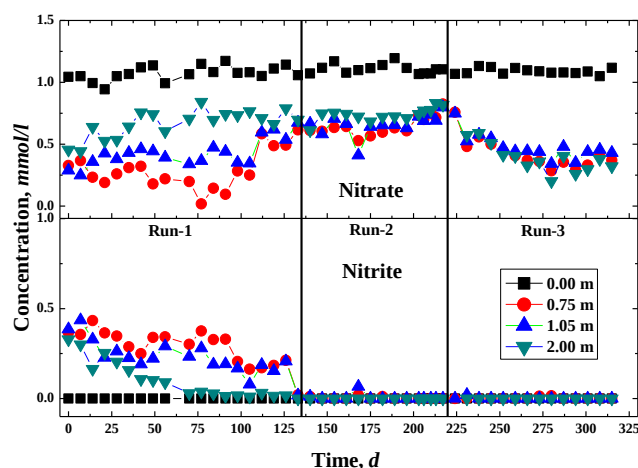


Fig. 2 Time courses of nitrate and nitrite

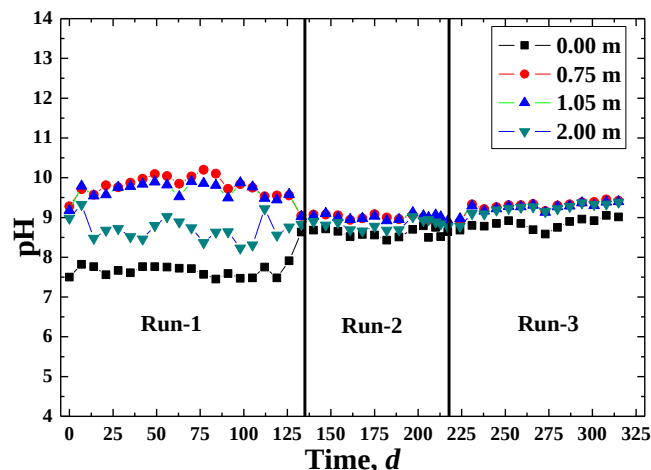


Fig. 3 Time courses of pH

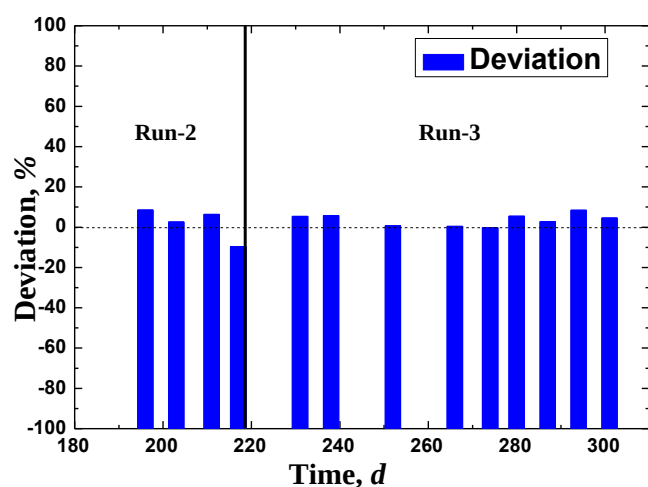


Fig. 4 Result of H₂ gas mass balance

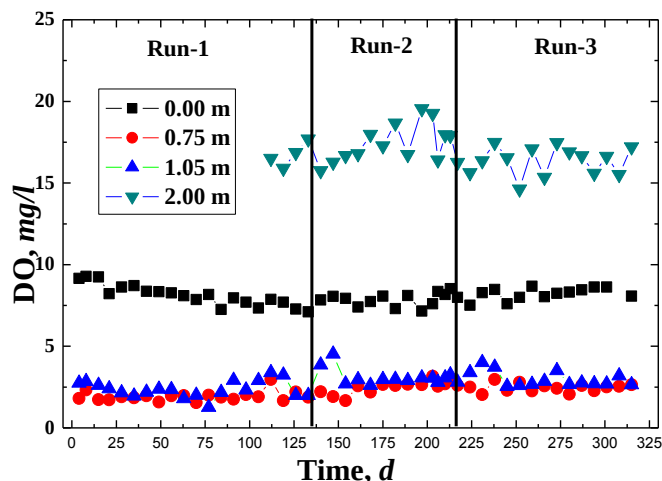


Fig. 5 Time courses of DO

4. CONCLUSIONS

From continuous experiments, it was demonstrated that stable denitrification and oxygenation/aerobic oxidation were achieved in the present *in-situ* process. A further long-term study under different H₂/O₂ injection conditions as well as kinetic analysis would be needed to evaluate the optimum design and operation conditions.

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