

Continuous Treatment of Groundwater by *In-Situ* Denitrification and Oxidation
 Process with Injection of Electrolytic Hydrogen and Oxygen
 Waseda University Student Member ○Jian Ye
 Waseda University Student Member Akane Kiga
 Waseda University Student Member Katsuki Naito
 Waseda University Regular Member Yutaka Sakakibara

1. INTRODUCTION

Nitrate contamination in groundwater due to over use of nitrogen fertilizer and/or inappropriate managements of livestock waste has become one of worldwide environmental issues. Several researches have been carried out to study the remediation of contaminated groundwater as a drinking water source. *In-situ* biological denitrification is one of the most promising processes, owing to relatively low cost, decomposition to a harmless product, nitrogen gas. In our former studies (Ye, J. *etc.*, 2013), the effectiveness of an *in-situ* process was demonstrated. Therefore, in this study, focusing on nitrate contaminated shallow aquifers, a long-term continuous experiments with injection of electrolytic hydrogen and oxygen were conducted under different nitrate loading conditions to evaluate the denitrification and oxygenation performances.

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). 18 sampling ports were allocated in horizontal and vertical directions with 300 mm interval, as shown in Fig. 1. Synthetic groundwater was fed and withdrawn continuously by using peristaltic pumps to realize a uniform flow. In experiment, hydrogen and oxygen gases were produced by using a solid polymer electrolyte (SPE) membrane electrode, and were injected using peristaltic pumps at the bottom of port 6 and port 15, respectively. Experimental conditions (Runs 1~3) were shown in Table 1.

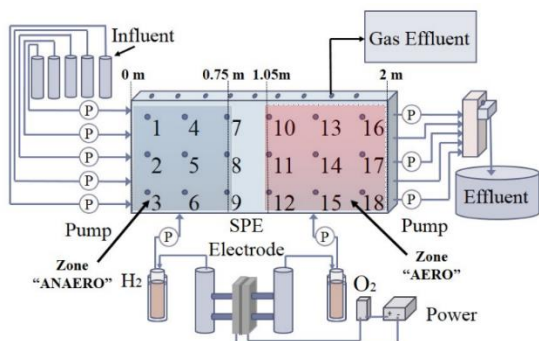


Fig. 1 Scheme of *in-situ* denitrification.

Table 1 Experimental conditions.

	Run 1	Run 2	Run 3
Flow rate (Q) (l/d)	≈ 5.8	≈ 11.5	≈ 23.0
HRT (d)	≈ 2.7	≈ 1.4	≈ 0.7
Temperature	20 ± 2 °C		
Inorganic Carbon (as C)	120-130 mg/l		
H ₂ gas (mmol/d)	≈ 15	≈ 27	≈ 42
O ₂ gas (mmol/d)	≈ 8	≈ 16	≈ 32

3. RESULTS AND DISCUSSION

Fig. 2 shows time course changes in nitrate and nitrite concentrations at positions of 0 m (influent), 0.75 m, 1.05 m, and 2 m (effluent) in Runs 1 to 3, respectively. In addition, pH values, DO, turbidity/chromaticity and TOC at the same positions were shown in Figs. 3 to 6, respectively. Concentrations as well as other values shown in all figures were average values of liquids obtained along three vertical sampling ports. In Runs 1 and 2, the nitrate removal rate kept very stable while almost no significant nitrite accumulation observed. In Run 3, the nitrate removal rate kept similar with Runs 1 and 2 while very small amount of nitrite accumulated in the hydrogen injected zone, but was oxidized to nitrate in a subsequent oxygen-injected zone. From Fig. 3 it could be concluded that after denitrification, the pH of the synthetic groundwater varied from weak acidic range to neutral.

Fig. 4 shows time course changes in dissolved oxygen. In runs 1 and 2, DO decreased to around 2 mg/l but in Run 3, DO only decreased to around 3 mg/l in hydrogen-injected zone. The reason was unknown, but it was supposed

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Contact address: Department of Civil and Environmental Engineering, Waseda University, 3-4-1 Okubo, Shinjuku Ku, Tokyo, 1698555

Tel/Fax: +81-3-5286-3902. E-mail: sakaki@waseda.jp

that because of the larger flow rate which lead to shorter reaction time in this zone. DO in all runs increased over 20 mg/l on average in oxygen-injected zone. Fig. 5 shows time course change of turbidity and chromaticity. Generally, these water qualities tended to decrease with distance of aquifer and became 0.2 NTU and 1.0 PCU in effluent. As glass beads have a poor adsorption capacity of these parameters, it was thought suspended solids as well as soluble and colloidal constituents exerting chromaticity were absorbed or attached to, or decomposed by biofilms formed on glass beads.

Fig. 6 shows time course changes of TOC and difference of TOC between influent and effluent. Deionized water used in this study contained about 2.6 mg/l TOC on average. Although data variations were observed, the TOC difference between influent and effluent was about -0.83 mg/l on average, indicating TOC produced in aquifer was removed. During the continuous experiments for totally over 700 days including precedent continuous experiments (Ye, J *et al*, 2013), no clogging was observed. Based on experimental results in Figs. 2 to 6, it can be concluded that the present in situ process has superior performances in terms of stability, effluent water quality, and simplicity in operation.

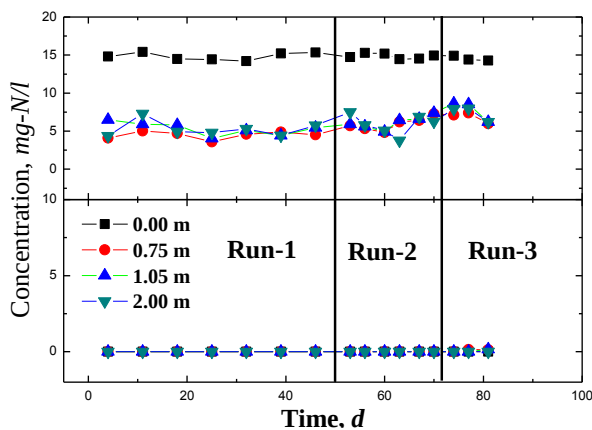


Fig. 2 Time courses of nitrate and nitrite.

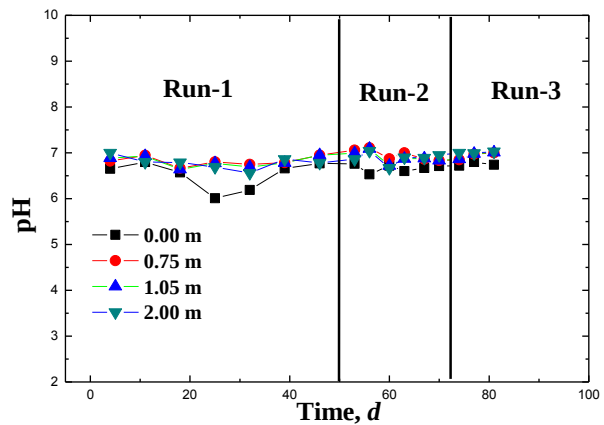


Fig. 3 Time courses of pH.

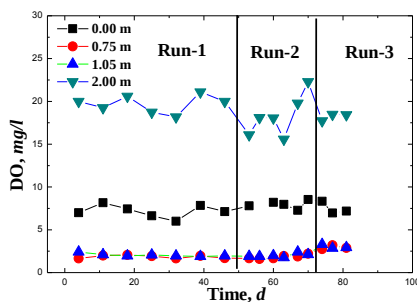


Fig. 4 Time courses of DO.

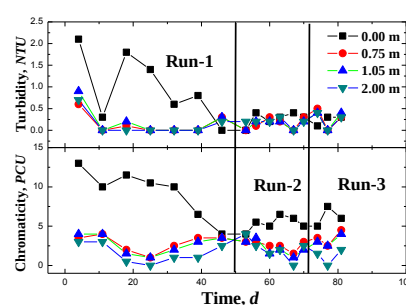


Fig. 5 Time courses of turbidity and chromaticity.

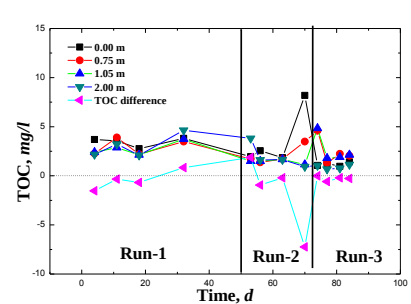


Fig. 6 Time courses of TOC.

4. CONCLUSIONS

From experimental results, the present in-situ process would be a feasible alternative to former biological denitrification process. A further study on more different operating/loardong conditions and a longer term study of its stability as well as kinetic analysis would be needed to evaluate the optimum design and operation conditions.

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