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Submerged bed versus unsaturated flow reactor: A pressurized hydrogenotrophic denitrification reactor as a case study

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HIGHLIGHTS

- Two modes of pressurized hydrogenotrophic denitrification reactor were compared.
- The unsaturated-flow mode with liquid recirculation presented higher rates and $k_L\alpha$.
- The unsaturated-flow mode with liquid recirculation presented lower effluent TSS.
- The operation under saturated-flow mode with gas recirculation was more stable.
- Energy consumption of gas recirculation is expected to be significantly lower.

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ABSTRACT

The paper compares the main features of a submerged bed reactor (SuBR) with bubbling and recirculation of gas to those of an unsaturated flow reactor (uSFR) with liquid recirculation. A novel pressurized closed-headspace hydrogenotrophic denitrification system characterized by safe and economic utilization of H_2 gas was used for the comparison.

Under similar conditions, denitrification rates were lower in the SuBR as a result of a lower effective biofilm surface area and overall gas-liquid mass transfer coefficient k_La. Similar values of effluent DOC were achieved for both reactors, although effluent suspended solids concentration of the SuBR were substantially higher. On the other hand, the required cleaning frequency in the SuBR was 2.5 times lower. Moreover, the SuBR is expected to reduce the recirculation energy consumption by 0.35 kWh/m^3 treated.

1. Introduction

Treatment processes of diverse water contaminants include submerged bed reactors and unsaturated flow biofilm reactors (similar to trickling filters). Trickling filters are characterized by simplicity, high degradation efficiency, low operation costs and small footprints. One of the main disadvantage of trickling filters is the risk of clogging with the accompanying need for frequent reactor cleaning (Daigger and Boltz, 2011; Eding et al., 2006; Epsztein et al., in press; Lekang and Kleppe, 2000). In submerged systems, the bubbles scour excess sludge from the carriers by the shear forces of the turbulent gas and, therefore, submerged systems are considered less susceptible to clogging than trickling filters (Schlegel and Koeser, 2007). Another drawback of trickling filters is the high hydraulic and energy-demanding recirculation commonly applied in order to achieve full media wetting (Eding et al., 2006; Epsztein et al., in press).

A novel pressurized reactor for hydrogenotrophic denitrification of groundwater operating at high denitrification rates together with minimal hydrogen loss and low risk was recently presented (Epsztein et al., 2016). The main novelty of the reactor is the operation under a pressurized closed headspace without any gas discharge. The common concern of N_2 gas build-up in a pressurized denitrifying system is addressed by the idea that in continuous operation a gas-liquid equilibrium is achieved according to Henry's law and the effluent water carries excess N_2 gas out of the reactor.

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Nomenclature

A	effective biofilm surface area [m^2/m^3]
a	specific interfacial area [m^2/m^3]
δ	length of gas-liquid boundary layer [m]
$D_{f,H}$	diffusion coefficient of H_2 in the biofilm [m^2/d]
$D_{f,N}$	diffusion coefficient of NO_3^- in the biofilm [m^2/d]
H^*	equilibrium concentration of dissolved H_2 [mg/L]
H_e	effluent concentration of dissolved H_2 [mg/L]
$k_{0f,H}$	degradation rate of H_2 in the biofilm [g/($L_{biofilm}\cdot$d)]
$k_{0f,N}$	degradation rate of NO_3^--N in the biofilm [g/($L_{biofilm}\cdot$d)]
k_La	overall volumetric gas-liquid mass transfer coefficient of H_2 [1/d]
N_e	effluent NO_3^--N concentration [mg/L]
N_i	inlet NO_3^--N concentration [mg/L]
$q_{\max,N}$	maximal specific degradation rate of NO_3^--N [g/(gVSS$\cdot$d)]
$q_{\max,H}$	maximal specific degradation rate of H_2 [g/(gVSS$\cdot$d)]
Q	volumetric flow rate [mL/min]
Q_R	liquid recirculation flow rate [mL/min]
r_H	overall H_2 degradation rate [g/(L$\cdot$d)]
r_N	overall NO_3^-—N degradation rate [g/(L$\cdot$d)]
V	reactor volume [L]
υ	stoichiometric mass ratio [g H_2/g NO_3^-—N]
X_f	biofilm density [gVSS/mL]

Since N_2 reaches equilibrium and is not accumulated over time, there is no need for gas discharge and the risky and economic H_2 loss to atmosphere through gas purging of the reactor is prevented. Hydrogen loss is therefore limited only to the dissolved H_2 in the effluent and H_2 utilization efficiencies above 92% were achieved. The operation under low-pressurized headspace consisting uniquely of H_2 and N_2 gases prevents hazardous H_2—O_2 contact and minimizes the risk of explosion in case of failure (Epsztein et al., 2016).

On top of the inherent advantages of safety and economics, the new reactor was designed to ensure high denitrification rates in comparison to existing hydrogenotrophic systems by using high-surface-area plastic carriers and maintaining high mass transfer of H_2 gas. The high mass transfer of H_2 gas can be accomplished by operating the reactor either under an unsaturated flow regime where water is recirculated through the H_2 gas-enriched headspace and trickled over the biofilm carriers (Epsztein et al., 2016), or with submerged bed where gas is recirculated from the headspace to the bottom and bubbled through the submerged bed.

The main objective of the current research is to compare between two types of biofilm reactors using the above hydrogenotrophic denitrification reactor as a case study: a submerged bed reactor (SuBR) and an unsaturated flow reactor (uSFR). The inherent features of the two reactors are discussed with a focus on the effective biofilm surface area and gas-liquid mass transfer.

2. Materials and methods

2.1. Experimental setup

The SuBR was based on the same reactor tank used for the uSFR (Epsztein et al., 2016) as shown in Fig. 1.

The comparison between the two reactors was based on the same packing volume of plastic carriers (total surface of 900 m^2/m^3, Aqwise). However, in order to allow for good mixing and fluidization of the carriers in the SuBR (as in fluidized or moving bed reactor), the carriers filling ratio chosen for the SuBR was ~60% (instead of 100% in the uSFR), and therefore a higher volume of the PVC cylindrical column (diameter of 10.5 cm) was utilized for the SuBR tests (height of 90 cm in the SuBR instead of 51 cm in the uSFR). The reactors were continuously fed with nitrate-contaminated groundwater. The level switches controlling water drainage were located in the reactor's bottom and top part for the unsaturated (i.e. uSFR) and saturated flow mode (i.e. SuBR), respectively. When enough liquid collected at the reactors and reached the level switch, a drainage valve was opened and a portion of treated water was released (i.e. pulsed discharge). A detailed description of the equipment in the uSFR was given earlier (Epsztein et al., 2016). The SuBR was connected to a gas supply (H_2 cylinder with pressure regulator), feed pump (Diaphragm pump model 7090-42, Cole-Palmer), gas recirculation pump (Peristaltic pump model 7553-75, Cole-Palmer), water recirculation pump (optional) (FL-2403, ProPumps) and pH controlling unit (standard pH electrode, pH controller – pH190, Alpha; hydrochloric acid tank and acid pump – gamma/L, ProMinent). Gas recirculation from the reactor's headspace was introduced at the bottom of the reactor through an aquarium-type air diffusion stone.

The feed solution for all experiments was tap water mixed with concentrated stock solutions of $NaNO_3$ and KH_2PO_4. The volumetric flow rate was 450 mL/min and effluent NO_3^--N was controlled by adjusting the inlet NO_3^--N concentration. Water temperature was maintained constant at 27.5 ± 1 °C. Bulk pH was kept at 7 ± 0.1 by dosing hydrochloric acid. Influent, effluent and water from the top part of the reactors were collected for further analyses. All rate calculations in this work were based on the packing volume of the carriers (i.e. $V = 4.4$ L).

2.2. Analytical methods

Nitrate was determined using a Metrohm 761 ion chromatograph (IC) equipped with a 150 mm Metrosep A Supp 5 column with column guard and suppressor using a CO_3^{-2}/HCO_3^- eluent. Nitrite-N and alkalinity were measured according to Standard Methods (Method 4500 and Method 2320, respectively). The total suspended solids (TSS) concentration was also carried out according to Standards Methods (APHA et al., 1995). Total Organic Carbon (TOC) concentration was determined by a TOC-VCPH analyzer (Shimadzu, Kyoto, Japan). DOC concentration was determined by performing TOC analysis on samples filtered through 0.22 mm syringe filter.

2.3. Reactors operation

According to the concept developed, normal reactors operation is done under a constant total pressure (i.e. pressure of H_2 and N_2 gases). In the beginning of the process the partial pressure of N_2 increases and the partial pressure of H_2 decreases till gas-liquid equilibrium is achieved. The partial pressures of H_2 and N_2 gases at equilibrium depend on the amount of NO_3^--N removed per litre of treated water (Epsztein et al., 2016).

In this work the operational conditions were changed according to the specifications of each experiment. When excess of H_2 and NO_3^--N was required, the operation to steady state was performed with the highest gas or liquid recirculation (1.5 and 8 L/min,