Journal of Environmental Management xxx (2017) 1–5

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Pressurized hydrogenotrophic denitrification reactor for small water systems

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ARTICLE INFO

Article history:
Received 4 January 2017
Received in revised form
23 February 2017
Accepted 3 March 2017
Available online xxx

Keywords:
Drinking water
Hydrogenotrophic denitrification
Hydrogen utilization
Nitrate removal
Nitrogen accumulation
Perchlorate removal

ABSTRACT

The implementation of hydrogenotrophic denitrification is limited due to safety concerns, poor H_2 utilization and low solubility of H_2 gas with the resulting low transfer rate. The current paper presents the main research work conducted on a pressurized hydrogenotrophic reactor for denitrification that was recently developed. The reactor is based on a new concept suggesting that a gas-liquid equilibrium is achieved in the closed headspace of denitrifying reactor, further produced N_2 gas is carried out by the effluent and gas purging is not required.

The feasibility of the proposed reactor was shown for two effluent concentrations of 10 and 1 mg NO_3^--N/L. Hydrogen gas utilization efficiencies of 92.8% and 96.9% were measured for the two effluent concentrations, respectively. Reactor modeling predicted high denitrification rates above 4 g NO_3^--N/($L_{reactor}$·d) at reasonable operational conditions. Hydrogen utilization efficiency was improved up to almost 100% by combining the pressurized reactor with a following open-to-atmosphere polishing unit. Also, the potential of the reactor to remove ClO_4^- was shown.

1. Introduction

The benefits of using H_2 gas as the electron donor for biological denitrification of groundwater were discussed previously. Among them, the clean nature and the low cell yield of hydrogenotrophic bacteria are the major advantageous features, resulting in small waste sludge production, minimal reactor clogging and reduced-cost post treatment. Additional advantage is the low price of H_2 gas compared to other electron donors per electron-equivalent delivered for contaminant reduction (two-thirds the price of methanol). Therefore, H_2 gas is an excellent choice for decentralized and small water systems where a simple and reliable technology with minimal manpower control is required. However, supplying H_2 gas at high transfer rates, yet economically and safely, remained the main challenge limiting the use of hydrogenotrophic denitrification (Karanasios et al., 2010). In some cases, a co-contamination of NO_3^- and ClO_4^- is observed and requires an overall solution for removing both ions (Zhao et al., 2014).

A summary of the prior technology available for hydrogenotrophic denitrification was given elsewhere (R. Epsztein et al., 2016a). Among these technologies, the membrane biofilm reactor (MBfR) has gained the most attention due to its safe and economic gas delivery system with close to 100% utilization efficiency of H_2 gas. Membrane fouling and scaling together with difficulties of biomass control are possible drawbacks of a typical MBfR (Karanasios et al., 2010). Additionally, the lower surface area provided by the membrane for biofilm growth compared to the surface area provided by plastic carriers in a packed- or fluidized-bed reactor may result in lower denitrification rates. For comparison, in the original paper presenting the MBfR, the specific total (i.e. clean) surface area reported was 390 m^2/m^3 (Lee and Rittmann, 2000), while Aqwise® plastic carriers, for example, provide a specific total surface area of 900 m^2/m^3. Despite the drawbacks described, the MBfR provides a promising solution for H_2 delivery and is implemented in full-scale in various groundwater treatment plants in California, USA since 2012 for the treatment of various pollutants (Martin and Nerenberg, 2012).

To the best of our knowledge, there are no earlier reports of hydrogenotrophic systems based on pressurized reactors with closed headspace. Most of conventional hydrogenotrophic denitrification reactors based on packed or fluidized bed present a similar H_2 delivery scheme of gas purging to the atmosphere in order to improve H_2 transfer rates and enable discharge of N_2 gas produced during denitrification. This operation, of course, results in

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http://dx.doi.org/10.1016/j.jenvman.2017.03.010
0301-4797/

a significant release of H_2 gas to atmosphere with its related economic and safety concerns.

A novel pressurized hydrogenotrophic denitrification reactor was recently proposed (R. Epsztein et al., 2016a). The reactor is based on a new concept suggesting that N_2 gas pressure reaches a constant level in the closed headspace of denitrifying reactor and therefore purging is not required (see explanation in the next section). The reactor is characterized by high denitrification rates, minimal hydrogen loss and low risk and can serve also for ClO_4^- removal. A following small open-to-atmosphere polishing unit can be used to eliminate residual dissolved H_2 and allow for safe effluent discharge. The simplicity of the new reactor may encourage its full implementation, especially in remote and small water plants, where process monitoring and control should be minimized.

In the following research work, the pressurized reactor's proof of concept and capability to operate at high denitrification rates and high H_2 utilization efficiencies are described. Also, investigation of the combined pressurized reactor-polishing unit treatment scheme and the potential ClO_4^- removal in the pressurized reactor are addressed.

2. Materials and methods

2.1. Explanation of the new concept

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 the effluent water carries excess N_2 gas out of the reactor. The dissolved N_2 concentration in the reactor reaches a constant level according to the concentration of NO_3^--N removed and therefore the partial pressure of N_2 in the reactor must also remain constant and correlate with the dissolved N_2 concentration according to Henry's law. For example, in the case of typical conditions of an inlet NO_3^--N concentration of 25 mg/L and a projected reactor effluent at drinking water regulations of 10 mg NO_3^--N/L, approximately all 15 mg NO_3^--N/L removed are converted to N_2 (assuming low biomass yield). An additional source of N_2 in the effluent is atmospheric N_2 dissolved in the influent water and carried into the reactor (~14 mg N_2/L under conditions of normal air mixture with 0.8 bar of atmospheric N_2 gas and Henry's constant of 17 mg N_2/[L·bar] at 20 °C). Therefore, the effluent water will contain about 29 mg N_2/L. Assuming for simplification that the reactor is completely mixed so the dissolved N_2 concentration is the same at any position in the reactor, this dissolved N_2 concentration will result in the development of a new gas-liquid equilibrium in the closed-headspace reactor with N_2 pressure around 1.7 bar according to Henry's law (29 mg N_2/L divided by 17 mg N_2/[L·bar]). Applying a total pressure of 2 bar, for instance, will leave room for 0.3 bar of H_2 gas at gas-liquid equilibrium. Since N_2 reaches equilibrium and does not further accumulate 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. 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.

In its original version presented in the current paper, the reactor is operated under an unsaturated flow regime as a trickling filter where water is recirculated and trickled over the biofilm carriers (Fig. 1). Plastic carriers with high surface area are used and together with high mass transfer of H_2 gas due to the unsaturated flow, high denitrification rates are achieved. The reactor is continuously fed with NO_3^--contaminated groundwater. When enough liquid collects at the reactor's bottom and reaches a level switch, a drain valve is opened and effluent water is released (i.e. pulsed discharge).

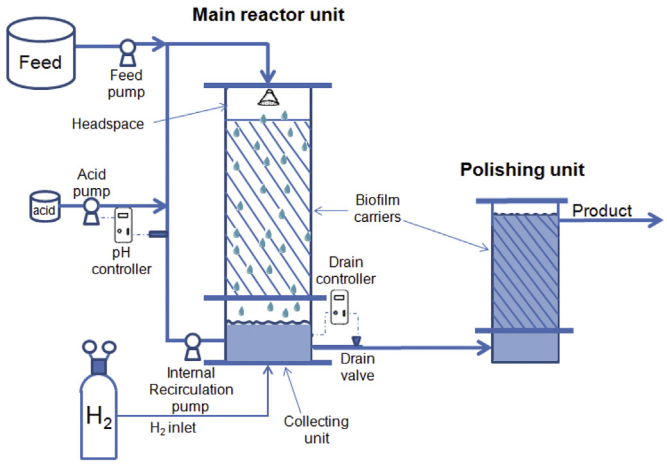

Fig. 1. Schematic diagram of the full experimental system.

An alternative version of the pressurized reactor, using a submerged bed where gas is recirculated from the reactor's headspace to the bottom and bubbled through the submerged bed, was tested in another work (R. Epsztein et al., 2016c).

2.2. Experimental setup

A schematic diagram of the full experimental system is illustrated in Fig. 1.

The full system included the main reactor unit and a following polishing unit to remove the residual H_2 in the reactor effluent. Except for the experiment testing the polishing unit performance (section 3.3), only the main reactor unit was used. The main reactor unit comprised of a clear PVC cylindrical reactor 100 cm in height and 10.5 cm in diameter divided into three unequal parts. The top part of the reactor (height 29 cm) served as an empty headspace, the middle part (height 51 cm) contained plastic biofilm carriers (total surface area of 900 m^2/m^3, *Aqwise*) and was separated by a metal screen from the bottom part (height 20 cm) of the reactor where recirculating water collected. The reactor was connected to a gas supply (H_2 cylinder with pressure regulator), feed pump (Diaphragm pump model 7090-42, Cole-Palmer), recirculation pump (FL-2403, ProPumps) and pH controlling unit (standard pH electrode, pH controller – pH190, Alpha; hydrochloric acid tank and acid pump – gamma/L, ProMinent).

In the experiment described in section 3.3, the main reactor unit was connected in a row to a PVC cylindrical polishing unit 22 cm in height and 10.5 cm in diameter, filled with the same plastic biofilm carriers as in the main reactor unit (see Fig. 1). The effluent water from the main reactor unit was introduced at the bottom of the polishing unit and released at the top part. The polishing unit was operated under a saturated-flow mode and its discharge was open to atmosphere.

For reactor start-up, a 2 L solution consisting of tap water enriched with NO_3^-, bicarbonate (added as 2 g of $NaHCO_3$) and phosphate with 0.5 L bacteria originating from a former hydrogenotrophic reactor was prepared and recirculated through the reactor in a batch mode under a constant H_2 pressure of 2 bar by purging. Water temperature was maintained constant at 25.5 ± 1 °C for the proof of concept trials (section 3.1). Model results (section 3.2) were based on model development experiments performed at

Please cite this article in press as: Epsztein, R., et al., Pressurized hydrogenotrophic denitrification reactor for small water systems, Journal of Environmental Management (2017), http://dx.doi.org/10.1016/j.jenvman.2017.03.010