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Research review paper

Biological nitrate removal from water and wastewater by solid-phase denitrification process

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ABSTRACT

Nitrate pollution in receiving waters has become a serious issue worldwide. Solid-phase denitrification process is an emerging technology, which has received increasing attention in recent years. It uses biodegradable polymers as both the carbon source and biofilm carrier for denitrifying microorganisms. A vast array of natural and synthetic biopolymers, including woodchips, sawdust, straw, cotton, maize cobs, seaweed, bark, polyhydroxyalkanoate (PHA), polycaprolactone (PCL), polybutylene succinate (PBS) and polylactic acid (PLA), have been widely used for denitrification due to their good performance, low cost and large available quantities. This paper presents an overview on the application of solid-phase denitrification in nitrate removal from drinking water, groundwater, aquaculture wastewater, the secondary effluent and wastewater with low C/N ratio. The types of solid carbon source, the influencing factors, the microbial community of biofilm attached on the biodegradable carriers, the potential adverse effect, and the cost of denitrification process are introduced and evaluated. Woodchips and polycaprolactone are the popular and competitive natural plant-like and synthetic biodegradable polymers used for denitrification, respectively. Most of the denitrifiers reported in solid-phase denitrification affiliated to the family *Comamonadaceae* in the class *Betaproteobacteria*. The members of genera *Diaphorobacter*, *Acidovorax and Simplicispira* were mostly reported. In future study, more attention should be paid to the simultaneous removal of nitrate and toxic organic contaminants such as pesticide and PPCPs by solid-phase denitrification, to the elucidation of the metabolic and regulatory relationship between decomposition of solid carbon source and denitrification, and to the post-treatment of the municipal secondary effluent. Solid-phase denitrification process is a promising technology for the removal of nitrate from water and wastewater.

Contents

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1. Introduction

Increasing levels of nitrate in receiving waters have been becoming serious issues worldwide due to the intensive application of fertilizers and pesticides, and sewage irrigation. The high concentration of nitrate has the potential risks of eutrophication and toxic algal blooms in receiving waters (Ghafari et al., 2008). Nitrate is identified as one of the hazardous contaminants in drinking water because it can cause infantile methemoglobinemia (blue baby syndrome) and reduction of nitrate into nitrites in saliva might induce the formation of nitrosamines, which are known carcinogens (Matiju et al., 1992). The maximum admissible concentration limit of nitrate in drinking water is 10 mg L^{-1} as nitrate-nitrogen (NO_3-N) set by the US Environmental Protection Agency and 50 mg L^{-1} as nitrate by World Health Organization to reduce the risks to human health (Tsai et al., 2004).

Biological nitrification and denitrification is a very important topic in the field of water pollution control (Wang and Yang, 2004; Liu et al., 2005; Chen et al., 2006). Different technologies have been researched and developed for nitrate removal, including ion exchange, adsorption, membrane separation, electrodialysis, chemical denitrification and biological denitrification (Aslan and Turkman, 2003; Wang and Kang, 2005). Biological denitrification is conducted by denitrifying microbes which use nitrate as terminal electron acceptor, and organic and inorganic substances as electron donor and energy source for sustaining the microbial growth (Ines et al., 1998; Ghafari et al., 2008). There are two types of biological denitrification, heterotrophic and autotrophic. Autotrophic denitrifiers utilize hydrogen, iron or sulfur compounds as energy source and inorganic carbon compounds such as carbon dioxide and bicarbonate as carbon source (Karanasios et al., 2010). Heterotrophic denitrifiers which use organic carbon compounds as carbon source are the most common denitrifiers in nature (Van Rijn et al., 2006). Heterotrophic biological denitrification is considered to be more economically, practically on a large scale, and ultimately reduce nitrate to nitrogen gas with high selectivity (Ovez et al., 2006a; Schipper et al., 2010b). The traditional technique is to add the water soluble substances such as methanol, ethanol, acetic acid and glucose into the denitrification reactor (Modin et al., 2007; Bill et al., 2009). There are the risks of insufficient doses or overdosing that entails a deterioration of the effluent quality. A complex process control and continuous monitoring is demanded. Moreover, some liquid carbon sources such as methanol and ethanol have security risks during storage, transportation and operation owing to their toxicity and inflammability. Recently, solid-phase denitrification which uses solid substances involving natural plants-like materials and synthetic biodegradable polymers served as carbon source for denitrification and biofilm carriers, has proved to be a promising alternative to remove nitrate from water and wastewater (Hiraishi and Khan, 2003; Boley and Muller, 2005; Chu and Wang, 2016).

This paper presents overview of the application of solid-phase denitrification for nitrate removal from water and wastewater. The types of solid carbon sources commonly used, the parameters affecting denitrification rate such as temperature and dissolved oxygen (DO), the characteristics and microbial community of biofilm attached on the biodegradable carriers, the adverse effects of solid-phase denitrification and the cost of denitrification are introduced and evaluated. The challenge of this technique and future outlook are proposed. The present paper would be useful for researchers and engineers in the field of nitrogen removal from water and wastewater.

2. Solid-phase denitrification: principle and characterization

Fig. 1 illustrates the reaction mechanism involved in solid-phase denitrification process. The polymers used in this process are called solid carbon source, which can be natural or synthetic, they should be water insoluble and biodegradable. The solid carbon source was initially hydrolyzed by extracellular enzymes such as lipase excreted by the microbes in attached biofilm and then decomposed into soluble and small-molecular substrates. Most of the substrates are utilized by denitrifying microbes to act as electron donor to reduce nitrate to nitrite, nitric oxide, nitrous oxide and finally to nitrogen gas, which is the most likely and the favorite pathway. There is still another route of dissimilatory nitrate reduction to ammonium (DNRA). DNRA competes with denitrification and converts nitrate to ammonium rather than converting nitrate to N_2 (Van Rijn et al., 2006). It has been reported that DNRA is a minor process involved in nitrate removal and less than 4–10% of removed nitrate was attributed to DNRA (Gibert et al., 2008; Healy et al., 2012). In addition, there is possibility that some of substrates are anaerobically digested to produce methane instead of denitrification. In the presence of oxygen, a part of substrates might be degraded by aerobic biodegradation process (Boley and Muller, 2005), in which CO_2 and biomass are produced.

Since the solid carbon sources are accessible by denitrifying microbes only after decomposition, the amount of the released organic carbon is regulated by bacteria responding to nitrate levels in the aqueous phase. Therefore, the risks of overdosing or insufficient dose could be avoided. The control and supervision of the process is simple (Gutierrez-Wing et al., 2012). Nowadays, solid-phase denitrification has been applied for in-situ groundwater remediation, for the treatment of drinking water, groundwater and wastewater with a low C/N ratio, for the tertiary treatment of the secondary effluent and recirculating aquaculture system for nitrate removal.

3. Solid carbon source commonly used for denitrification

3.1. Types of solid carbon sources and their denitrification rate

There are two kinds of solid carbon sources available for solid-phase denitrification: the natural plant-like materials and synthetic biodegradable polymers. Table 1 presents the types of carbon sources, their advantages and drawbacks. The natural materials such as woodchips, straws and cottons are cheap and available, but the high release of dissolved organic carbon (DOC) and color was found in the effluent, especially during the start-up period (Volokita et al., 1996; Aslan and Turkman, 2004; Ovez, 2006; Xu et al., 2009; Robertson, 2010; Cameron and Schipper, 2012). The biodegradable polymers, including polyhydroxyalkanoate (PHA), poly-3-hydroxybutyric acid (PHB), poly-3-hydroxybutyrate-*co*-hyroxyvelate (PHBV), polycaprolactone (PCL), polybutylene succinate (PBS) and polylactic acid (PLA), are proved to be the suitable carbon sources for denitrification due to their low release of DOC, however their cost are relatively high (Honda and Osawa, 2002; Hiraishi and Khan, 2003; Walters et al., 2009; Zhao et al., 2009; Zhou et al., 2009; Shen and Wang, 2011; Takahashi et al., 2011; Wu et al., 2013b).

The carbon sources used as an electron donor for denitrification have influence on the conversion rate of nitrate to nitrogen. Table 2 illustrates the nitrate removal efficiency and denitrification rate using different

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