Chemical Engineering Science 152 (2016) 488–496

Contents lists available at ScienceDirect

Chemical Engineering Science

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

Model-based evaluation on simultaneous nitrate and arsenite removal in a membrane biofilm reactor

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HIGHLIGHTS

- A novel approach to simultaneous NO_3^- and As(III) removal in MBfR was proposed.
- Kinetics of autotrophic As(III) oxidation process was determined.
- A biofilm model coupling DAMO and autotrophic As(III) oxidation was developed.
- MBfR performance and microbial structure were assessed under different conditions.
- The optimal CH_4 supply was found inversely proportional to the corresponding HRT.

ARTICLE INFO

Article history:
Received 1 April 2016
Received in revised form
29 May 2016
Accepted 20 June 2016
Available online 21 June 2016

Keywords:
Membrane biofilm reactor
Nitrate removal
Arsenite removal
Denitrifying anaerobic methane oxidation
Mathematical modeling

ABSTRACT

Nitrate (NO_3^-) and arsenite (As(III)) are two major contaminants in groundwater, which could cause significant risks to human wellbeing and ecological system. In this work, a single-stage membrane biofilm reactor (MBfR) coupling denitrifying anaerobic methane (CH_4) oxidation (DAMO) and autotrophic As(III) oxidation processes was proposed for the first time to achieve the in-situ or ex-situ simultaneous removal of NO_3^- and As(III) from groundwater. CH_4 is supplied to the MBfR through gas-permeable membranes while NO_3^- and As(III) are provided in the bulk liquid. A mathematical model was developed by integrating the well-established biokinetics of DAMO microorganisms with the kinetics of As(III)-oxidizing bacteria (AsOB). The key parameter values of AsOB were specifically estimated using the batch experimental data of an enriched pure AsOB culture in conjunction with thermodynamic state calculations. The maximum specific growth rate of AsOB (μ_{AsOB}) and the yield coefficient for AsOB (Y_{AsOB}) were determined to be 0.00161 h^{-1} and 0.016 g COD g^{-1} As, respectively. The modeling results demonstrated that both influent surface loading (or hydraulic retention time (HRT)) and CH_4 surface loading played important roles in controlling the steady-state microbial community structure and thus significantly affected the system performance. The As(III)/NO_3^- ratio between 0.1 and 2 g As g^{-1} NO_3^--N in the influent would have no significant impact on the overall system performance despite the varying microbial composition in the biofilm. Through properly adjusting the influent surface loading (or HRT) and CH_4 surface loading whilst maintaining a sufficient biofilm thickness at a suitable influent As(III)/NO_3^- ratio, the maximum removal efficiencies of total nitrogen and As(III) could both reach above 95.0%, accompanied by a high CH_4 utilization efficiency of up to 99.0%.

1. Introduction

The remediation of contaminated groundwater represents a significant issue which needs to be addressed urgently, considering the potable use of groundwater by over 50% of the global population (Jadhav et al., 2015). Arsenic (As) has been recognized as a crucial pollutant present in groundwater, with arsenite (As(III)) and arsenate (As(V)) being the predominantly occurring species. Arsenic has been classified as a human carcinogen by the United States Environmental Protection Agency (EPA), and arsenicosis could arise via ingestion of As-containing water and its subsequent accumulation in the body (Jadhav et al., 2015). Though the permissible limit of As in water is recommended at 10 μg L^{-1} by the World Health Organization (WHO), the actual As contamination level in groundwater varies greatly and could reach up to 300 mg L^{-1} (Mukherjee et al., 2006), depending on the specific contamination type and source of the site. A number of technologies have been established and applied to treat As-laden

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http://dx.doi.org/10.1016/j.ces.2016.06.049

groundwater, including the injection of nitrate (NO_3^-) (Harvey et al., 2002). However, as a matter of fact, NO_3^- itself serves as another important pollutant commonly observed in groundwater. Due to the widespread application of nitrogen-containing fertilizers as well as the discharge of industrial and municipal wastewaters, the NO_3^- concentration in groundwater has been remarkable (Della Rocca et al., 2007), although its level varies in different places. The maximum acceptable contamination level of NO_3^- has been documented at 10 mg N L^{-1} by EPA (Yang and Lee, 2005), while it's set/recommended at 11.3 mg N L^{-1} by European Union countries and the WHO (Zhu and Getting, 2012). Nevertheless, the current NO_3^- contamination level in the groundwater of the United States and some European countries has been reported to increase to 200 mg L^{-1} (Chen et al., 2014a). Excessive NO_3^- not only favours the eutrophication of receiving waters (Fennessy and Cronk, 1997; Koren et al., 2000) but also causes serious health problems, such as methemoglobinemia (Knobeloch et al., 2000). Therefore, the potential in-situ or ex-situ integration of As and NO_3^- removal would be fairly desirable for groundwater with joint or separate contamination of As and NO_3^-, respectively.

The discovery of denitrifying anaerobic methane (CH_4) oxidation (DAMO) microorganisms (Raghoebarsing et al., 2006) and autotrophic As(III)-oxidizing bacteria (AsOB) (Oremland et al., 2002; Rhine et al., 2006) offers a potential solution to the above-mentioned problem by means of implementing biological treatment processes. Through utilizing CH_4 as the electron donor, DAMO archaea are capable of reducing NO_3^- to nitrite (NO_2^-) (Haroon et al., 2013) while DAMO bacteria are able to convert NO_2^- to nitrogen gas (N_2) (Ettwig et al., 2010). AsOB can use the energy and reducing power from As(III) oxidation for carbon dioxide (CO_2) fixation and cell growth under both aerobic and anaerobic NO_3^--reducing conditions (Zhang et al., 2015a). The oxidized As(V) produced is generally regarded as less mobile in the environment and less toxic to organisms than As(III) (Sun et al., 2010). Through proper introduction of CH_4 in conjunction with the creation of a favorable reactor configuration, the coculture of DAMO microorganisms and AsOB could be facilitated to likely achieve the simultaneous removal of NO_3^- and As(III) from groundwater with various contamination conditions.

Attached growth (i.e., biofilm) has been considered particularly suitable for retaining sufficient biomass of microorganisms with slow growth kinetics, such as DAMO archaea and DAMO bacteria (Chen et al., 2014b; Winkler et al., 2015). To ensure a high transfer and utilization efficiency of externally supplied CH_4, the membrane biofilm reactor (MBfR) has been commonly applied/proposed to couple DAMO with other processes in a single-stage system, with high achievable removal performance (Chen et al., 2014b, 2015; Shi et al., 2013). Therefore, a novel MBfR integrating DAMO and autotrophic As(III) oxidation is proposed hereby for the

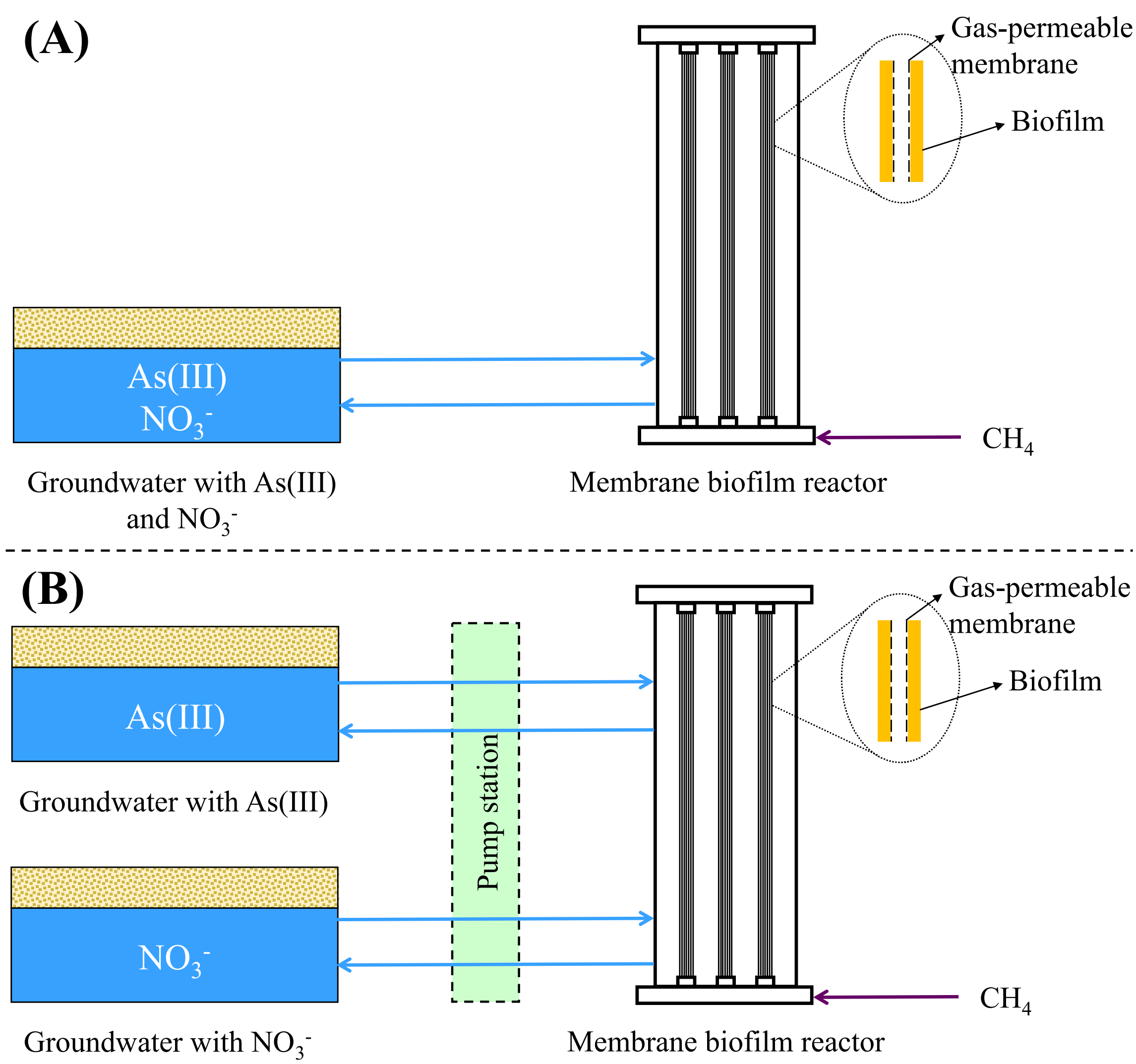

Fig. 1. The conceptual application of the MBfR to achieve (A) the in-situ remediation of groundwater contaminated with NO_3^- and As(III) simultaneously and (B) the ex-situ treatment of the combined stream of groundwater polluted by NO_3^- and As(III) separately.