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Potential of hydrolyzed polyacrylamide biodegradation to final products through regulating its own nitrogen transformation in different dissolved oxygen systems☆

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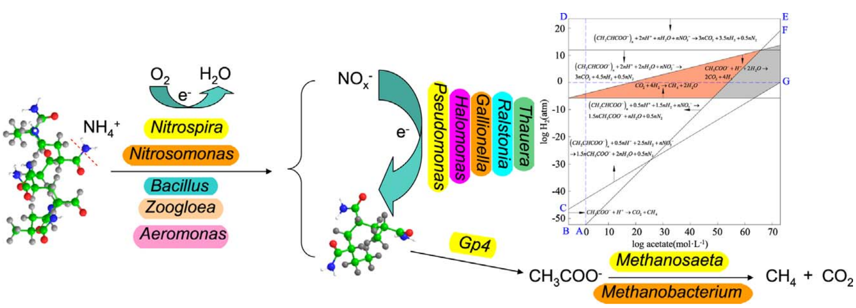
GRAPHICAL ABSTRACT



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ABSTRACT

Potential of hydrolyzed polyacrylamide (HPAM) biodegradation to final products was studied through regulating its own nitrogen transformation. Under the conditions of 2, 3 and 4 mg/L of DO, HPAM removal ratio reached 16.92%, 24.51% and 30.78% and the corresponding removal ratio reached 49.15%, 60.25% and 76.44% after anaerobic biodegradation. NO_3^--N concentration was 9.43, 14.10 and 17.99 mg/L in aerobic stages and the corresponding concentration was 0.17, 0.07 and 0.008 mg/L after anaerobic biodegradation. Oxygen as electron acceptors stimulated the activities of nitrification bacteria and other functional bacteria, thus further enhanced nitrification and HPAM biodegradation. NO_3^- (from HPAM oxidation) as electron acceptors stimulated the activities of nitrate-reducing, acetate-producing and methanogenic microorganisms and they could form a synergistic effect on denitrification and methanogenesis. Thermodynamic opportunity window revealed that NO_x^- could accelerate anaerobic HPAM bioconversion to methane. Aerobic and anaerobic growth-process equations of cells verified that the metabolism on HPAM was feasible.

1. Introduction

With the global energy demands increasing, offshore oil exploration and development have developed rapidly. Polymer flooding technology has been the main force to improve oil recovery in Bohai sea oil fields (Zhao et al., 2018). Organic pollutants, including hydrolyzed polyacrylamide (HPAM) and oil, caused by offshore oil development may have potential risk on marine ecological environment and human health. Disposal of HPAM-containing wastewater has been a difficult problem to be solved in offshore oil exploration and marine

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environmental protection. The disposal of offshore organic pollutants has also been listed in the outline of marine business and science and technology development plan of China, and regarded as one of seven priorities in the 13th five-year plan for science and technology innovation in the area.

Various kinds of methods have been involved in the treatment for HPAM-containing wastewater, such as chemical degradation (Ramsden and McKay, 1986; Gu et al., 2016), physical disposal (Deng et al., 2002; Zhang et al., 2013), biodegradation (Bao et al., 2010) and the combination of these methods (Lu and Wei, 2011). Biodegradation and bioconversion technologies, which are both environmentally friendly and cost effective, have been applied to treat HPAM-containing wastewater (Shpiner et al., 2009; Guezennec et al., 2015).

The participation of electron acceptors plays a significant role in regulating pollutant bioconversion. Additional electron acceptors added into bioreactors to facilitate the removal ratios of pollutants have been investigated by researchers. The biodegradation performance of pollutants is different with the participation of various kinds of electron acceptors, which include O_2, NO_x^-, SO_4^{2-} and so on (Schmidt et al., 2017). NO_x^- as an important electron acceptor is responsible for the biodegradation of many kinds of pollutants. Sabumon (2009) explored the effect of NO_x^- and SO_4^{2-} as electron acceptors on ammonia oxidation under anoxic condition and found that additional electron acceptors could enhance the removal of total nitrogen. In addition, additional NO_3^- as electron acceptors could promote anoxic pyridine biodegradation (Shen et al., 2015). NH_4^+ could be produced via biological HPAM hydrolysis. However, whether it is possible for NH_4^+ oxidized to NO_x^- by nitrification bacteria to further facilitate HPAM biodegradation is still unknown. In addition, the process of nitrogen shift is unclear during the biodegradation of HPAM as the sole nitrogen source and electron donor.

The potential of HPAM biotransformation is different in different oxygen environments. Although anaerobic microorganisms were common in oil reservoirs, a significant proportion of aerobic microorganisms was present in coal seams and oil sands of subsurface (An et al., 2013). Adequate and detailed investigations about the potential of aerobic and anaerobic HPAM bioconversion in subsurface are still absent. Waki et al. (2018) employed two activated sludge systems and modeled a useful tool to optimize the biodegradation of swine wastewater with different dissolved oxygen (DO) conditions and found that it was possible to remove nitrogen efficiently with a low-concentration DO. In addition, different electron acceptors (e.g., SO_4^{2-} and Fe^{3+}) were involved in anaerobic HPAM bioconversion and they could regulate the methanogenesis in the process of HPAM biotransformation (Zhao et al., 2018). However, insufficient research has been targeted in HPAM biodegradation with potential electron acceptors produced from its own rather than by adding extra electron acceptors. The effect of oxygen and potential electron acceptors on the regulation mechanisms of aerobic and anaerobic HPAM bioconversion was still not clear. In addition, the feasibility of aerobic and anaerobic HPAM bioconversion to final products needs to be further explored.

Thus, the objectives of this study were to (1) investigate the removal efficiencies of HPAM and the biotransformation of nitrogen with O_2 as electron acceptors under aerobic systems and NO_x^- as electron acceptors under anaerobic systems following the corresponding aerobic stages, (2) explore the regulation mechanisms of functional microorganisms on HPAM and nitrogen biotransformation, (3) clarify the feasibility of aerobic and anaerobic HPAM bioconversion to final products from the view of thermodynamics and (4) construct growth-process equations of microbial cells on HPAM in the whole systems.

2. Materials and methods

2.1. Experimental set-up

The Experimental systems in this study were designed by coupling an aerobic biofilm reactor (AeBR) and an anaerobic baffle reactor (AnBR). Three groups of this pattern were employed. Each AeBR with an identical effective volume of 68 L was made of organic glass (Yan et al., 2016). The fixed-film fillers made of polyester silk were distributed in each AeBR evenly to promote the formation of biofilm. In addition, Air compressor was placed in the bottom of each AeBR to maintain the dissolved oxygen (DO) concentration to 2 mg/L, 3 mg/L and 4 mg/L, respectively. As for AnBR systems, each reactor had four compartments with the capacity of 8.5 L (Zhao et al., 2016). The hydraulic retention time (HRT), sludge retention time (SRT) and temperature were set at 24 h, 30 d and 30 °C, respectively. The influent was fed to the AeBR via a peristaltic pump and fully contacted with the biofilm and activated sludge. After the aerobic process, the supernatant flowed into AnBR for further anaerobic degradation.

2.2. Wastewater ingredients

HPAM sample with the average relative molecular mass about 2.2×10^7 purchased from Changan Polymer Group Company (Dongying, China). It could perform as energy source for aerobic and anaerobic microbial growth because it contained both carbon and nitrogen element. In addition, it was partly hydrolyzed due to the existence of the amide group. Some microelements were employed to ensure microbial growth and metabolism (Luo et al., 2008). $NaHCO_3$ was applied to balance the influent pH ranging from 7.0 to 7.5. The specific ingredients of influent water were shown below (mg/L): HPAM, 300; $KH_2PO_4{\cdot}3H_2O$, 50; Na_2SO_4, 30; $CaCl_2$, 25; $CuCl_2{\cdot}5H_2O$, 5; $NiCl_2{\cdot}6H_2O$, 5; $ZnCl_2$, 5; $MnCl_2{\cdot}4H_2O$, 5; $CoCl_2{\cdot}6H_2O$, 5; H_3BO_3, 5; $AlCl_3$, 2.5.

2.3. Bioreactor inocula and domestication

The oilfield sludge contained functional bacteria for aerobic and anaerobic HPAM biodegradation was obtained from Zhan Three oil production plant (Dongying, China) and employed as the inoculated sludge for sludge domestication of each reactor. During the start-up stage, the HPAM concentration of influent water increased from 0 mg/L to 300 mg/L and the corresponding glucose concentration of influent water decreased from 300 mg/L to 0 mg/L with the gradient of 50 mg/L. When the removal ratios of HPAM, TOC and COD reached a stable level at a gradient concentration in each AeBR-AnBR, the next domestication gradient was carried out. Each domestication stage lasted for about 10 days. The start-up stage lasted for 70 days and the whole systems achieved a steady state after that. Mixed liquor suspended solids (MLSS) concentration was maintained about 4000–5000 mg/L. The influent, effluent of AeBR and effluent of AnBR were acquired every day. Mature sludge samples were collected from biofilm in AeBR and anaerobic activated sludge in AnBR. All samples were frozen in a refrigerator at −20 °C for further analysis.

2.4. Analytical methods

All water samples were filtered through 0.45 μm microporous membranes for detection. HPAM concentration was measured by the starch-cadmium iodide method (Bao et al., 2010). The COD and TOC values were obtained by a COD digester (DRB200, HACH, America) and a TOC analyzer (Multi N/C, Analytikjena, Germany) respectively. The pH was determined by a digital pH meter (pB-10, Sartorius Group, Germany). The concentrations of mixed liquor suspended solids (MLSS), NH_4^+-N, NO_3^-, NO_2^- were measured according to the standard methods (APHA, 2005). Acetate concentration was measured via High Performance Liquid Chromatography (HPLC) (Alliance e2695, Waters Science and Technology Co., Ltd., Shanghai, China). Injection volume, flow rate and column temperature were set to 20 μL, 0.7 mL/min and 55 °C, respectively. The wavelength of UV–vis detector was 210 nm and 0.05% phosphoric acid was employed as the mobile phase (Zhao et al., 2018). CH_4 and CO_2 productions were measured through