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Experimental and modeling study on nitric oxide removal in a biotrickling filter using *Chelatococcus daeguensis* under thermophilic condition

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HIGHLIGHTS

- Biotrickling filter inoculated with *C. daeguensis* TAD1 for NO removal is achieved under aerobic condition at 50 °C.
- Mathematical model method is employed to describe the detailed mechanisms for NO removal in BTF.
- Gaseous chemical oxidation for NO removal is coupled which is not considered in present BTF models.

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ABSTRACT

In this study, the development of a thermophilic biotrickling filter (BTF) system to inoculate a newly isolated strain of *Chelatococcus daeguensis* TAD1 for the effective treatment of nitric oxide (NO) is described. It was successfully started up in 35 days and effectively removed NO from the oxygen contained simulated gas at 50 °C. A mathematical model based on the mass transfer in gas–biofilm interface and chemical oxidation in gas phase was developed. Steady-state experimental data under different inlet NO concentration and empty bed retention time (EBRT) condition were used to verify the proposed model. The model can well reproduce the experimental results and the sensitivity analysis demonstrates that the model is not dependent on the accuracy of the parameters excessively.

1. Introduction

Increasing emission of nitrogen oxides (NOx) from various industrial processes and transportation activities, especially coal-fired power plants, are widely regarded as risks to global environment and human health (Barnes et al., 1995). Nitric oxide (NO), the major NOx component (Nagase et al., 1997; Wang et al., 2006), is a precursor for tropospheric ozone depletion and the main constituent in photochemical smog (Chen et al., 2006; Gao et al., 2011). It may also react with moisture in the air to form nitrous acid, which has been implicated in acid rain (Akimoto, 2003). Although the production of NOx can be significantly reduced by combustion process control, post-combustion flue gas treatment is required to satisfy the current regulatory air standards (Wang et al., 2006). Therefore, technologies to remove NO from the effluent gas have attracted wide attention. Conventional post-combustion technologies for NOx removal include selective catalytic reduction, selective non-catalytic reduction, adsorption, scrubbing, and so forth (Chen et al., 2009). All these are effective methods, but they often suffered from some problems, such as the catalysts are easily poisoned and hazardous wastes should be disposed (Barnes et al., 1995; Bögner et al., 1995).

Bioprocesses including biotrickling filter (BTF) and biofilter are emerging post-combustion control technologies that can be used as potential alternatives for NOx containing dilute gases purification (Chen and Ma, 2006; Ramírez et al., 2009). The principle of BTF is to treat the gaseous pollutants in a packed bed of damp material on which pollutant-degrading microorganisms are attached. According to Ligy and Marc, chemolithoautotrophic organisms such as *Thiobacillus denitrificans* can reduce nitric oxide (NO) to nitrogen gas under anoxic conditions in the biotrickling filter (Philip and Deshusses, 2003). Several studies have also proven the possibility of NO removal in bioreactors at ambient temperatures. Jiang et al. have successfully isolated a new strain of *Pseudomonas putida* SB1 and achieved effective treatment for nitric oxide at ambient temperature (Jiang et al., 2009). Jun et al. have used a bench-scale anaerobic rotating drum biofilter (RDB)

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to demonstrate its ability to treat off-gas containing nitric oxide and found the optimal temperature was around 30 °C (Jun et al., 2008). Yang et al. have demonstrated that nitric oxide could be treated by a bench-scale biofilter at environmental temperature and NO removal efficiency was inversely proportional to inlet NO and O_2 concentration (Yang et al., 2007). However, it is noted that many industrial emission gas are very high in temperature. For example, in coal-fired power plants, influent flue gas stream for a NOx removal reactor is the emissions from a sulfur dioxide scrubber, and temperature of gas exiting from the scrubbers of the effluent is still between 50 and 60 °C. Therefore, the BTF process which employs mesophilic microorganisms to remove NOx cannot be directly used in such a high temperature condition. In this case, pre-cooling process is necessary before the BTF process, which will increase the investment and operation cost significantly.

In order to solve this problem, very few investigators have conducted on isolating and applying some thermophilic microorganisms for NOx removal, which may offer great savings and greatly extend the applicability of BTF. Flanagan et al. (2002) have studied on the treatment of NO by microorganisms at 53 °C and oxygen-free environment (Flanagan et al., 2002). Lee et al. (2001) have pointed out that NO removal could be achieved in compost biofilters operated at 55 °C, but nitric oxide removal dropped to between 10% and 20% when oxygen was present in the influent stream. However, generally the flue gas vent out from the coal-fired power plants include not only NO but also O_2 (3–8%) (Lee et al., 2001). Thus, it is meaningful to study on NO removal by microorganisms under thermophilic and aerobic conditions. Up to now, there is scarcely report about biological treatment of NO under simultaneous aerobic and high temperature environment. Besides, theoretical modeling studies regarding NO removal in BTF reactor are very limited and present literatures about BTF modeling are concentrated on volatile organic compounds treatment (Cáceres et al., 2012; Song and Kinney, 2002; Zarook and Shaikh, 1997). It is also important that present reports about NO biological removal models have not taken gaseous chemical oxidation of NO elimination into consideration (Chen et al., 2009).

In the current study, a series of continuous experiments in a bench-scale BTF reactor have been carried out to examine the effect of *Chelatococcus daeguensis* TAD1 on NO thermophilic removal and to explore the operation parameters on reactor performance under 8% oxygen. In order to better understand and serve for scale up design of the BTF reactor, a mathematical model originated from the interface mass transfer balance has been developed to predict the relationship between operation conditions and reactor efficiency.

2. Methods

2.1. Microbe

In authors' previous study, a strain *C. daeguensis* TAD1 has been successfully isolated under high temperature and aerobic environment from the biofilm of an on-site BTF at the Ruiming coal-fired power plant (Guangzhou, China) (Liang et al., 2012; Yang et al., 2012). The 16S rRNA sequences (1385 bp) of *C. daeguensis* were examined in blastn (NCBI, USA) for similarities. The sequence data of the isolated strain has been submitted to the DDBJ/EMBL/GenBank databases under accession No. HM000004.

2.2. BTF media

The trickling nutrient liquid included the following contents: disodium succinate, 5.0 g L^{-1}; $Na_2HPO_4{\cdot}7H_2O$, 3.0 g L^{-1}; KH_2PO_4, 1.5 g L^{-1}; $MgSO_4{\cdot}7H_2O$, 0.1 g L^{-1}; NaCl, 4.7 g L^{-1}; trace element solution, 2 mL L^{-1}; pH 7–7.5. The trace element solutions for the bacterial growth consisted of the following components: EDTA, 50.0 g L^{-1}; $ZnSO_4$, 2.2 g L^{-1}; $CaCl_2$, 5.5 g L^{-1}; $MnCl_2{\cdot}4H_2O$, 5.06 g L^{-1}; $FeSO_4{\cdot}7H_2O$, 5.0 g L^{-1}; $(NH_4)_6Mo_7O_2{\cdot}4H_2O$, 1.1 g L^{-1}; $CuSO_4{\cdot}5H_2O$, 1.57 g L^{-1}; $CoCl_2{\cdot}6H_2O$, 1.61 g L^{-1}; pH 7.0.

2.3. Experimental setup and operation

A bench scale BTF system shown in Fig. 1 was used in this study. The BTF was constructed with cylindrical plexiglass. The height and diameter of the reactor was 50 and 8 cm, respectively. Porous ceramic particles (size 2–4 mm, bulk density 1.10 g cm^{-3}) were used as packing material. The packing space was at the height from 10 to 40 cm calculated from the bottom of the BTF. The out surface of the reactor was wrapped with heat tape which is covered with a layer of fiberglass insulator. The heat tape was controlled by a digital temperature controller using a thermo-sensor inserted into the BTF as an input signal. The reactor temperature was maintained at 50 ± 1 °C.

Inlet waste gas was synthesized by pure NO, N_2 and O_2 which were stored in respective cylinders. Three gases were mixed in the humidifier before entering the BTF. Different inlet NO concentration was controlled by adjusting the flow rate of each gas entering in the humidifier. Simulated NOx gas was humidified and preheated by passing through the humidifier. The humidifier was located in a thermostatic water bath (50 ± 1 °C). Just before the mixed gas entering the reactor, the flow rate and gas concentration were recorded. The constant volume (3 L) liquid reservoir for nutrients recycling was also controlled at 50 ± 1 °C by a thermostatic water bath. Nutrient liquid from the liquid reservoir was pumped and sprayed from upper surface of the packing materials to maintain adequate moisture and to provide the necessary nutrients for the growth of microorganisms. In order to avoid alien bacteria, all appliances and materials used in this experiment were pre-sterilized. Besides, the inlet and outlet gases were passed through sterile 0.45 μm bacterial air vents, and all trickling liquid was sterilized before using.

2.4. Analytical methods

For the thermophilic removal of NO in the continuous gas-streams experiment, the flux of NO, N_2 and O_2 was controlled by a mass flow controller (FM310-MT, OPINE, China). The NO, NO_2, and O_2 concentrations at both the inlet and outlet were analyzed by a flue gas analyzer (TESTO 350Pro, Germany) at 10 h intervals. The NO concentration at the inlet and outlet of the BTF were measured to determine the NO removal efficiency. Microbial counts were done in duplicate on DM/nitrate agar medium plates. A dilution series of biofilm was prepared in 8.5 g L^{-1} NaCl for plates incubated at 50 °C.

2.5. Model development

2.5.1. Basis of the model

A mathematical model has been developed for the present BTF reactor packed with uniform ceramic particles. *C. daeguensis* pure-culture biofilm is growing on the surface of the pelletized packing material under thermophilic condition. No free liquid is recirculated through the packing materials and nutrient liquid is continuously supplied using a top sprayer to minimize the effect of nutrients limitation on biofilm growth. Therefore, it can be considered that the overall BTF processes take place in two phases of gas and biofilm linked by a thin interfacial layer between the two phases. The influent polluted gas flows upward through the packed layer; at the gas-biofilm interface, dissolution equilibrium is achieved and the gaseous and interfacial liquid concentrations