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A New Approach for Removal of Nitrogen Oxides from Synthetic Gas-streams under High Concentration of Oxygen in Biofilters

Shao Bin HUANG*, Ju Guang ZHANG, He Ping HU, Yue SITU

Department of Applied Chemistry, South China University of Technology, Guangzhou 510640

Abstract: The potential of using denitrifying and nitrifying concurrent biofilters for the removal of nitrogen oxides from synthetic gas streams was studied under the condition of high oxygen concentration. It was found that more than 85% of nitric oxide was removed from synthetic combustion gas-streams which contained 20% oxygen and 350 μL/L NO, with a residence time of 60 seconds. In the process, it was found that the existing of oxygen showed no evident negative effect on the efficiency of nitrogen removal.

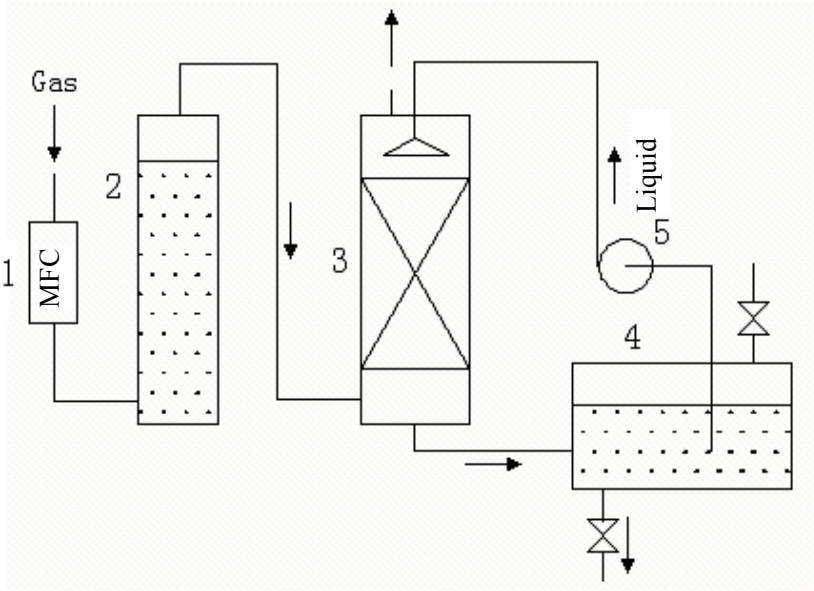
Keywords: Biofilter, nitrogen oxides, nitrification denitrification, aerobic.

Biofiltration has been employed for the treatment of volatile organic compounds, ammonia or hydrogen sulfide. It shows sound performance in these processes. Using biofilter to treat nitrogen oxides (NO_x) containing emissions has been studied in recent years. Under the anoxic or anaerobic conditions, it has been proved that NO_x is reduced to nitrogen gas efficiently in biofilter[1]. Oxygen showed remarkable inhibit on denitrifying action in the previous studies. For instance, biofilter was able to remove NO at level greater than 50% on oxygen-free condition, but below 20% efficiency could get with 2% oxygen at inlet gas-stream[2]. In reverse, oxygen redounds to nitrification. Some researchers built up an aerobic biofilter in which NOx was turned into NO_2^- or NO_3^- by autotrophic nitrite nitrobacterium. In that study, 41~52% of NO was removed with the influent NO concentration of 107.14 mg/m^3 and an empty-bed resident time of 3.5 minutes[3].

In this paper, a new approach for nitrogen removal in aerobic biofilter was introduced. Synthetic off-gas comprised of NO, O_2, N_2. N_2 was taken as the carrier gas. The flowing rate of gases was controlled by mass flux control. The concentration of NO, NO_2, O_2 was analyzed by flue gas analyzer (Quintox KM9106, Kane-May Company). Nitrate and nitrite were analyzed by an ion chromatography (DX-500, Dionex Corporation). Separation and elution of anions were carried out on an IonPac AS11 analytical column (4×250 mm) and IonPac AG11 guaranteed column (4×50 mm), utilizing 3.5 mmol/L Na_2CO_3 and 1.0 mmol/L $NaHCO_3$ as eluent with the velocity of 1.0 mL/min.

* E-mail: chshuang@scut.edu.cn

Figure 1 Sketch of biofiltration

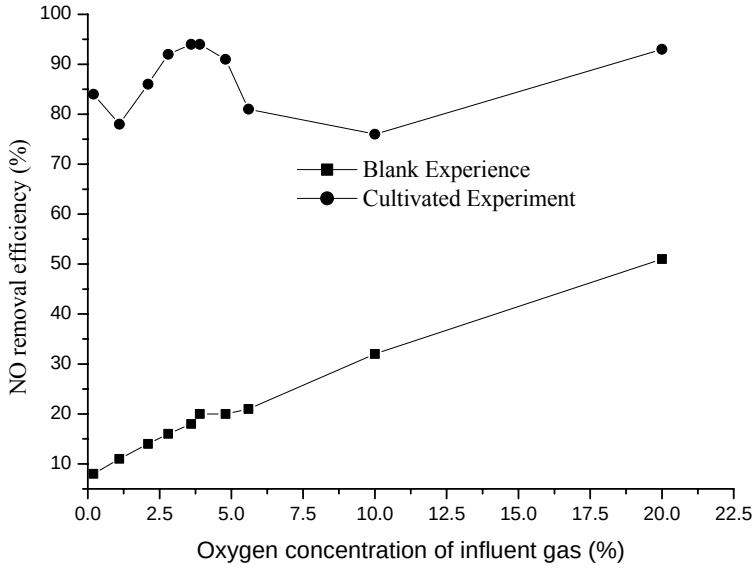


The system of biofiltration including: 1. mass flux control; 2. humidified column; 3. bioreactor; 4. nutrient vessel; 5. pump

The biofilter was constructed using a 40 cm length × 8 cm i.d. glass process pipe. Ceramic balls (ϕ 4 ~ 6 mm) was used as medium inoculated with post-domesticated denitrifying bacteria and nitrifying bacteria. Around the glass process pipe of biofilter was a fiber glass coating with a heating equipment installed to maintain the biofiltration temperature at the level of 50~53 ℃. Before entering the bioreactor, gases were passed through a humidified column to adjust of the humidity. Then synthetic gas-streams were directed into the biofilter from the bottom and let off from the top. A pump was used to supply biofilter with nutrients and moisture from the top of the bioreactor. The nutrient solution contained carbon resources, phosphate buffer, sodium chloride, and some volume of microelements. Glucose**,** lactic acid, broth and sodium acetate can be used as carbon resources, respectively. Part of leachate was collected into a vessel, where it was refreshed with new nutrients and recycled to reactor. Process of biofiltration is showed in **Figure 1**.

In order to explore the microbial nitrogen loss, a blank experiment was developed with sterilized medium. NO removal efficiency showed in **Figure 2** with different oxygen concentration at a resident time of 60 seconds. Before oxygen merged into influent gas-streams, less than 10% of NO was absorbed by packed materials. As the oxygen increased, there was a linear increase of NO removal efficiency. About 50% of NO was removed when 20% oxygen existing. The reason is that more NO was oxidized in high oxygen condition and transferred into liquid from the gas streams.

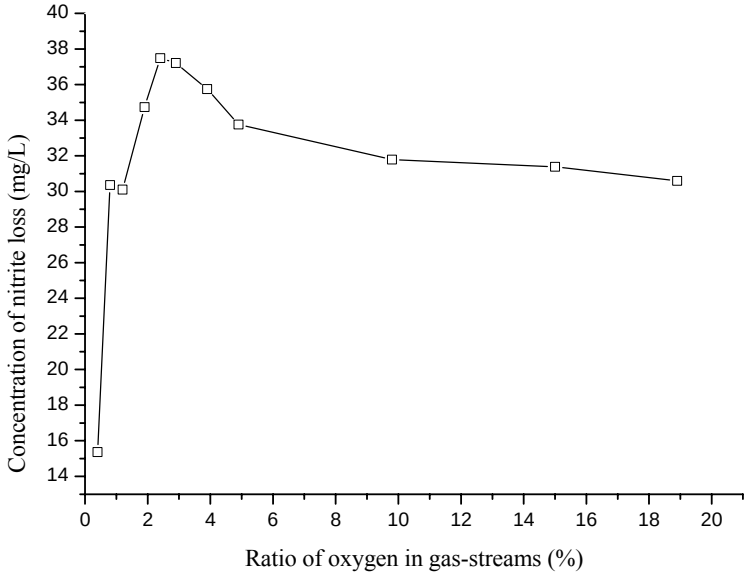
Figure 2 NO removal efficiency under different concentration of O_2



In the cultivated experiment, well cultivated denitrifying and nitrifying multi-species microbes were used for the experiment. The biofilter operated in anoxic condition at the beginning. Oxygen of different concentration was not mixed with the gas streams before the NO removal efficiency reached more than 85%. It was discovered that trace concentration of nitrate and nitrite emerged when more than 3% of oxygen injected into gas-streams, but accumulation of nitrate and nitrite never occurred during this process. A removal efficiency peak at 3.5% of oxygen (see **Figure 2**) verified both nitrification and denitrification to be active.

The mechanism of nitrogen loss can be imagined as that absorption and nitrification was the pretreatment of denitrification, whereby NO was turned to NO_2, NO_2^-, NO_3^-. Almost all of NO_2, NO_2^-, NO_3^- were finally turned to nitrogen and biomass during denitrification, but the fluctuation of the curve in **Figure 2** indicated that the transformation mechanism of NO in biofilter was more complex than imagined. Even though no nitrite and nitrate were added to nutrient liquid, about 1~5 mg/L nitrite can be detected in the leachate. So we can conclude that nitrifying action occurred in this bioreactor. But nitrite and nitrate did not accumulate in the recycle liquid, even the bioreactor has run for a long time. In order to verify this phenomenon of nitrogen loss in aerobic bioreactor, nitrite as a substitute for nitrogen oxides is used as the reactant, because nitrite can be nitrified and denitrified by microbes as well. During the biofiltration, nitrogen and oxygen in different ratio were aerated into the bioreactor. The efficiency of nitrite removal was showed in **Figure 3**.

Figure 3 Concentration of nitrite loss in different ratio of oxygen



Similar to nitrogen oxide removal, oxygen promoted the transfer of nitrite and a peak of nitrite disappeared near 3% oxygen. Nitrate less 1 mg/L put to the test, and no building-up of nitrate in the recycle liquid.

The biofilter could keep an efficiency of over 85% for more than 20 days with 20% of oxygen and a residence time of 60 seconds.

In a conclusion, well cultivated denitrifying and nitrifying multi-species bacteria were successfully used for the NOx removal in aerobic biofilter. The concentration of oxygen is an important factor, but high concentration of oxygen was found not to markedly inhibit process of nitrogen loss.

Acknowledgments

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