Int. J. Environ. Res., 5(3):779-786, Summer 2011
ISSN: 1735-6865

A performance Study of Simultaneous Microbial Removal of NO and SO_2 in a Biotrickling-Filter Under Anaerobic Condition

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Received 25 Aug. 2010; Revised 7 Oct. 2010; Accepted 20 Nov. 2010

ABSTRACT: Behaviors of simultaneous removal of NO and SO_2 using the coculture of anaerobic denitrifying bacteria and sulfate reducing bacteria were investigated in a bench-scale biotrickling-filter. Approximate 20 days were required to use the landfill leachate as the microbial seed to develop the biofilm on the surface of packing material. When the combined NO/SO_2 removal biotrickling-filter was operated at an empty bed residence time (EBRT) of 76 s and the NO and SO_2 feed concentrations of 2 and 2 g/m^3 respectively, the SO_2 removal efficiency was always above 95%, while NO removal exhibited an evident periodicity of 5-6 days for the initial 60 days after the attachment phase and then a steady-state NO-removal efficiency of around 90% was obtained after 130 days of continuous operation. Contrast experimental results indicated that the coculture in the combined NO/SO_2 removal biotrickling-filter showed a higher resistance to shock NO-loadings and a better tolerance of starvation than the single denitrifying bacteria in the NO removal biotrickling-filter.

Key words: Simultaneous removal of NO and SO_2, Coculture, Anaerobic denitrifying bacteria, Sulfate reducing bacteria, Biotrickling-filter

INTRODUCTION

The removal of nitrogen oxides (NOx), consisting of ±95% nitric oxide (NO) and ± 5% nitrogen dioxide (NO_2) (Flanagan *et al.,* 2002), and sulfur oxides (SO_2) emitted from stationary combustion facilities has been recently required to protect the atmospheric environment (Sada *et al.,* 1980). So far, a number of integrated processes for simultaneous removal of NO and SO_2 have been developed, such as electron beam treatment (EBA), pulsed corona induced plasma chemical process (PPCP), combination of flue gas desulfurization (FGD) plus selective catalytic reduction (SCR) and absorption. The absorbents include activated carbon (Zhu *et al.,* 2005), CuO/γ-Al_2O_3 (Jeong and Kim, 2000), Ca-pellets+K/Carbon-pellets (Bueno and Garcia, 2005), etc. EBA technology, although capable of obtaining 90% of SO_2 and 80% of NO removal efficiencies, is limited in practical use due to the high-energy consumption for producing energetic electrons from a large-capacity electronic accelerator (Liu *et al.,* 2007). Based on the same removal mechanism, PPCP reduces the energy consumption required for producing the energetic electrons or plasma by replacing the costly electronic accelerator with a pulsed high-voltage power supply, but some problems still exist, e.g., continuous addition of NH_3, relatively higher temperature of 65-100! and matching problem between the pulse power and the reactor (Wang *et al.,* 2009). Likewise, the FGD-SCR combination has not been widely used due to its large space demand, high water consuming, production of by-product gypsum and advanced wastewater treatment required before discharging (Zhao *et al.,* 2005). With respect to the absorption technique, a major disadvantage is the cost associated with the replacement or regeneration of the exhausted carbon and other absorbents (Hutchinson and Robinson, 1990). Therefore, there has been an increasingly urgent need for a simple, low-cost and single-stage treatment used for simultaneous removal of SO_2 and NO.

Simultaneous bioremoval of SO_2 and NO is considered as a promising alternative technique due to the reasons of low-temperature, low-consumption, low-cost, simplicity of operation and no secondary pollution. As early as 1991 Sublette et al. proposed the concept of combined removal of SO_2 and NO using the mixed cultures of *Desulfovibrio desulfuricans* (a sulfate-reducing bacteria) and *Thiobacillus denitrificans* (an autotrophic denitrifying bacteria) (Lee and Sublette, 1991). They also demonstrated that simultaneous removal of SO_2 and NO could be realized with the coculture of sulfate-reducing bacteria (SRB) and anaerobic denitrifying bacteria (ADB) (Dasu *et al.,* 1993), but they didn't further investigate the

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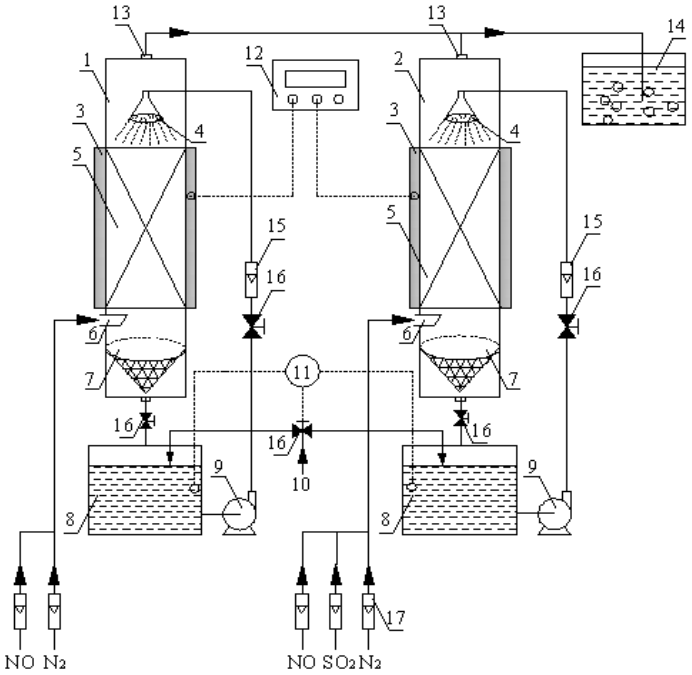
combined removal performance under different conditions. And then Philip and Deshusses utilized the activated sludge as the inoculum instead of specific bacteria strains to make the operation economically viable (Philip and Deshusses, 2003), however, they more focused on the complete treatment of SO_2 and ignored the combined removal behaviors. In the present study, simultaneous removal of SO_2 and NO using the coculture of SRB and ADB in a bench-scale biotrickling-filter was described. Both SRB and ADB were enriched from the landfill leachate which was initially inoculated into the filter. A comprehensive study on the long-term bioremoval performance was conducted under the selected operating conditions. In order to compare the NO removal performance, a parallel test using the single denitrifying bacteria to remove NO-rich gas was also carried out.

MATERIALS & METHODS

A schematic of the experimental setup is shown in Fig.1. The NO removal biotrickling-filter (NRBF) and the combined NO/SO_2 removal biotrickling-filter (CRBF) were operated in parallel to make a comparison of NO-removal performance between the single denitrifying bacteria and the coculture. Each biotrickling-filter (BF) was made of acryl glass. The total height of each BF was 200 cm with an inner diameter of 8 cm and a bed depth of 150 cm. And each BF was filled with the polyethylene Cascade Rings (25×13×1 mm; specific surface area of 228 m^2/m^3; bulk density of 65 kg/m^3). Additionally, each one was operated at a constant temperature (30 ± 0.5°C) with the heating tape which was controlled by a digital temperature controller (AI-708P, China). The pH of the recirculation liquid in each reservoir was controlled at 7.0±0.1 via a pH controller (Knick model 761, Germany) that automatically added 5 wt% $NaHCO_3$ solution to the liquid. The gas was fed to each BF from the bottom, while the nutrient liquid was supplied at a rate of 100 L/h from the top. SO_2, NO and N_2 contained in the feed gas were supplied from the compressed gas cylinders and then mixed in the gas mixer before entering the filter (not shown in Fig.1). The fluxes of SO_2, NO and N_2 were controlled by the gas flow meters. In each biotrickling-filter, the carrier gas (N_2) flow rate was controlled to keep a constant empty bed residence time (EBRT) of 76 s.

The landfill leachate used for seeding the filter was obtained from a municipal landfill in Tianjin, China. Prior to being filled to the filter, the packings were soaked in the landfill leachate for one week to facilitate



1-NO removal biotrickling-filter; 2-combined NO/SO_2 removal biotrickling-filter; 3-heating tape; 4-liquid distributor; 5-packings; 6-gas inlet; 7-preventing clogging net; 8-recycle liquid reservoir; 9-pump; 10-$NaHCO_3$ solution; 11-pH controller; 12-temperature controller; 13-gas outlet; 14-gas absorber; 15-liquid flow meter; 16-valve; 17-gas flow meter

Fig. 1. Schematic diagram of experimental setup

bacterial attachment to the surface of the packing materials.Then each BF filled with the pre-soaked packing materials was supplied with 1 L of landfill leachate and 10 L of respective selective medium (as seen in Table 1). Neither nitrate nor sulfate was contained in the medium to acclimate the microorganisms to NO or SO_2 as a terminal electron acceptor for ADB or SRB. In order to ensure the filter anoxic, each BF was flushed with N_2 until the gas phase had been exchanged 20 times (Beller *et al.,* 1996). And then we started the pumps and made them working continuously under anoxic conditions. For each BF, 1/2 of the liquid was replaced weekly by the fresh medium for the initial 20 days and then the whole liquid was removed and replaced weekly by an equal volume of fresh medium for the next 196 days, because the substrates of ammonium, thiosulfate and lactic acid were basically depleted within a week. The continuous measurements were conducted between 8:00 and 18:00 everyday. The specific operating conditions are shown in Table 2.

The concentrations of NO, NO_2, SO_2 and H_2S in the inlet as well as in the outlet were determined by a flue gas analyzer (KANE940 Multi-Gas Emissions Analyser, UK). The concentrations of nitrate, nitrite, sulfate, sulfite, total sulfide (dissolved H_2S, HS^-, S^{2-}) and the amount of protein were determined by a water quality analyzer (Merck NOVA 60 Spectroquant, Germany). The concentration of lactic acid was determined using a commercial lactic acid assay kit (Nos 330-1, Sigma). The number of SRB cells was measured using the fluorescence in situ hybridization (FISH) method with the SRB385 probe. The number of ADB cells was measured by combined use of stable-isotope probing (SIP), full-cycle rRNA analysis and FISH (Ginige *et al.,* 2005).

Table 1. Medium composition of anaerobic denitrifying bacteria (A) in NRBF and coculture (B) in CRBF

Substrates	A (g/L)	B (g/L)
NH_4Cl	1	1
KH_2PO_4	0.05	0.05
K_2HPO_4	0.2	0.2
thiosulfate	5	0
lactic acid	0	3.5
ascorbic acid	0	0.1
yeast extract	0	1
$NaHCO_3$	2.52	2.52
$FeSO_4 \cdot 7H_2O$	0.05	0.05
trace element*	0.02	0.02

* Trance element: Cu^{2+}, Co^{2+}, Mn^{2+}, Zn^{2+}, Ca^{2+} and ammonium molybdate

Table 2. Operation sequence and conditions

Day	Description	Feed gas of NRBF	Feed gas of CRBF	
		NO (g/m^3)	NO (g/m^3)	SO_2 (g/m^3)
1-20	Attachment phase	0.50-2.02	0.49-2.05	0.45-2.07
21-200	Long-term acclimation	1.99-2.05	2.01-2.08	2.02-2.04
201-202	NO-shock loadings	a	a	2.03-2.06
203-204	Recovery from shock loadings	2.03-2.05	2.00-2.06	2.01-2.08
205-210	Starvation-survival	0	0	0
211-216	Recovery from starvation	2.01-2.04	2.01-2.05	2.02-2.06

[a] On Day 201 and Day 202, NO feed concentrations on 1.5th and 5.5th hour of each day increased sharply to approximate 4.9 g/m^3.

RESULTS & DISCUSSION

The aim of this study is to develop a biological treatment for simultaneous removal of NO and SO_2 using a single biotrickling-filter. A 216-day continuous experiment was conducted to investigate the performance for simultaneous removal of NO and SO_2. In order to compare NO-removal stability, the ability of CRBF to resist to performance deterioration (i.e., NO-shock loadings and no gas feeding) was compared with NRBF.

Simultaneous removal of NO and SO_2 can be achieved using the coculture of anaerobic denitrifying bacteria and sulfate reducing bacteria in an attached biofilm bioreactor under anaerobic condition. In this process SO_2 was first reduced to H_2S by SRB, and then H_2S served as an electron donor for the reduction of NO to N_2 in denitrification by ADB. Sulfate produced from H_2S oxidation by ADB in turn served as an electron acceptor for the growth of SRB.

It's found that SRB obtained the competitive growth advantage prior to ADB and then ADB gradually became co-dominant with SRB (described in detail later in 3.2), which is inconsistent with the previous observation that heterotrophic denitrifying bacteria (hDB) can outcompete SRB for organic electron donor under anaerobic condition (Gevertz *et al.,* 2000; Hubert and Voordouw, 2007). Heterotrophic denitrifying bacteria didn't outcompete SRB probably due to the absence of nitrite, because nitrite is a strong inhibitor of SRB and hence considered as the key factor for successful competition of hDB for common organic electron donor (Hubert and Voordouw, 2007). Moreover, continuous H_2S production by SRB and insufficient organic carbon caused autotrophic denitrifying bacteria to dominate over heterotrophic denitrifying bacteria. Thus, H_2S produced by SRB served as the primary electron donor for NO reduction by ADB, which is well consistent with the experimental results discussed in 3.4. Furthermore, the coculture was acclimated to utilizing NO and SO_2 as the terminal electron acceptor by the long-term exposure to a gas mixture of NO, SO_2 and N_2 (carrier gas) and so the capacity of coculture for simultaneous removal of NO and SO_2 can be improved gradually with time.

For the initial 20 days, the concentration of NO (or SO_2) in the feed gas was increased stepwise to acclimate ADB (or SRB) to utilizing NO (or SO_2) as the terminal electron acceptor. A biofilm was developed on the surface of the packing material shortly after the startup of CRBF. The color of the attached biofilm in the CRBF gradually became dark brown and off-gas had a strong hydrogen sulfide odor after approximately 6 to 7 days, indicating the successful enrichment of fast-growing SRB from the landfill leachate. These observations were consistent with the FISH-analysis that SRB of which number was 1.5×10^9 cells/mg prot accounted for at least 80% of the coculture. During the next fortnight, however, the color gradually faded to light brown and the odor gradually disappeared, and meanwhile the outlet concentration of SO_2 was always below 100 mg/m^3. Estimates from FISH-analysis of the fractions of SRB and ADB were 50% (±15%) and 40% (±15%), respectively. All the above indicated that the coculture of SRB and ADB was successfully enriched from landfill leachate. By contrast, the time required for the development and acclimatization of ADB in NRBF was 7 to 9 days longer than that of CRBF, probably because SRB helped to shorten the startup time of ADB in the CRBF.

After this attachment phase, a 180-day (Day 21 to Day 200) continuous measurements of removal efficiencies of NO and SO_2 were initiated, which are shown in Fig. 2. It can be conclude that when both SO_2 and NO were fed at a constant rate of 2 g/m^3, the SO_2 removal efficiency was always above 95%, while NO removal exhibited an evident periodicity of 5-6 days for the initial 60 days (Day 21 to Day 80) after the attachment phase. However, the periodic fluctuation range gradually reduced until a steady-state removal of NO was reached after 130 days (Day 130 to Day 180) with the NO-removal efficiency of around 90%.

From a typical cycle as shown in Fig. 3, it's found that the daily N-removal ($NO+NO_2$, NO_2 is a result of oxidation of partial NO by trace oxygen existing in N_2) efficiency showed first a progressive increase and then decrease with time. This phenomenon was particularly obvious in the last 3 days of the cycle. During the first 3 days of the cycle, NOx ($NO+NO_2$) was removed at a relatively high efficiency, and the average daily N-removal efficiencies were 82.8%, 88.4% and 70.4%, respectively (see Fig.3.A, B and C). On the 4^{th} and 5^{th} days, the efficiencies dropped abruptly to below 40% or even approaching zero (see Fig.3.D and E). However, the removal efficiency on the 6^{th} or 7^{th} day was restored to above 82% miraculously (see in Fig. 2).

The possible reason for the daily curve-trend removal as well as long-term periodic removal may be explained as followed. Nitric oxide reductase, a comparatively very active enzyme, was first induced by the high NO feeding to maintain NO at such low concentrations that the potential toxicity of NO was not realized (Goretski *et al.,* 1990), so N-removal efficiency gradually increased. However, continuous exposure to such high NO environment caused the decrease in activity of nitric oxide reductase and hence N-removal efficiency reduced. After the physiological regulation of bacteria the higher NO removal efficiency resumed again. Furthermore, the bacteria' tolerance of

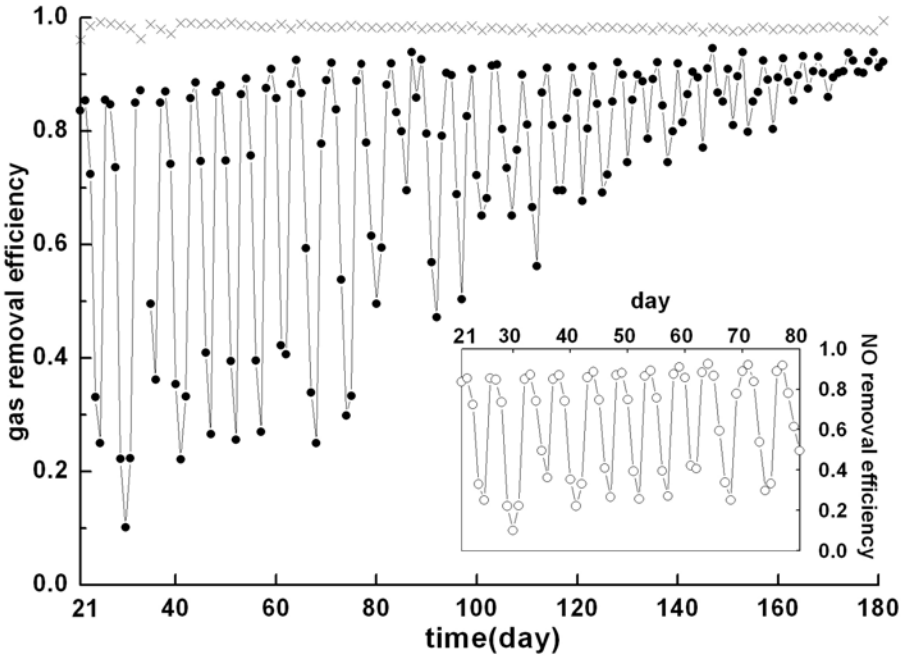


Fig. 2. Long-term experiments on NO removal efficiency (●) and SO_2 removal efficiency (×) as a function of time, and NO removal efficiency during the initial 60 days after the attachment phase (o)

NO toxicity was gradually enhanced by the long-term acclimatization and so the periodic fluctuation gradually disappeared.

The NO removal performances of NRBF and CRBF in response to NO-shock loadings and starvation were evaluated (Fig.4). When the NO inlet concentrations increased to 4.6-4.9 g/m^3 from 2 g/m^3 on the 1.5th and 5.5th hour of Day 201 and Day 202, the NO outlet concentrations of CRBF suddenly increased to 1.9-2.7 g/m^3 from 0.3 g/m^3 and then declined to the normal level within 4.5 hours after the NO inlet concentration returned to 2 g/m^3. Likewise, the NO outlet concentrations of NRBF returned to the normal level (around 0.5 g/m^3) within 4.5 hours after the 1st shock loading. But after the 2nd and 3rd shock loading, NRBF was not capable of resuming the normal efficient removal of NO within 4.5 hours and the average NO-removal efficiencies dramatically dropped from 61.3% to 41.6% and 31.5% respectively. After the 4th shock loading, the time required for NRBF to resume the normal NO-removal efficiency was approximate 21 hours which was nearly 5 times longer than that for CRBF.

When the high NO-removal efficiency was resumed after NO-shock loadings, gas was stopped to introduce into each BF from Day 205 to Day 210 to evaluate the tolerance of a 6-day starvation, which is shown in Fig.5. The coculture of CRBF could rapidly resume the high NO-removal efficiency within 20 hours immediately after starvation, whereas the single denitrifying bacteria of NRBF showed a poor NO-removal performance for the initial 20 hours after starvation and then resumed its normal removal efficiency after 60 hours.

These results indicated that the ability of CRBF to resist performance deterioration under NO-shock loading or starvation was superior to NRBF.The possible reasons for superiority of CRBF may be include (1) the alkali produced by SRB can balance the acid produced by denitrifying bacteria to stabilize the pH-value in the coculture of CRBF (Baalsrud and Baalsrud, 1954), (2) the poise of optimal redox potentials of denitrifying bacteria in CRBF might be prolonged with the aid of SRB (Bailey and Beauchamp, 1971), and (3) heterotrophic SRB contained in CRBF is able to produce slime-like materials that can help in adhering biofilms to the packing surfaces under long-term shear stress (Chou and Lin, 2000).

For simplification, H_2S is assumed to be the sole electron donor for NO reduction to nitrogen by the autotrophic denitrifying bacteria (ADB) under the anaerobic condition. The equations involved in the process are as followed:

$$2SO_2 + C_3H_6O_3 \xrightarrow{SRB} 2H_2S + 3CO_2 + H_2O \quad (1)$$

$$4NO + H_2S \xrightarrow{ADB} 2N_2 + SO_4^{2-} + 2H^+ \quad (2)$$

$$3SO_4^{2-} + 2C_3H_6O_3 \xrightarrow{SRB} 3H_2S + HCO_3^- \quad (3)$$

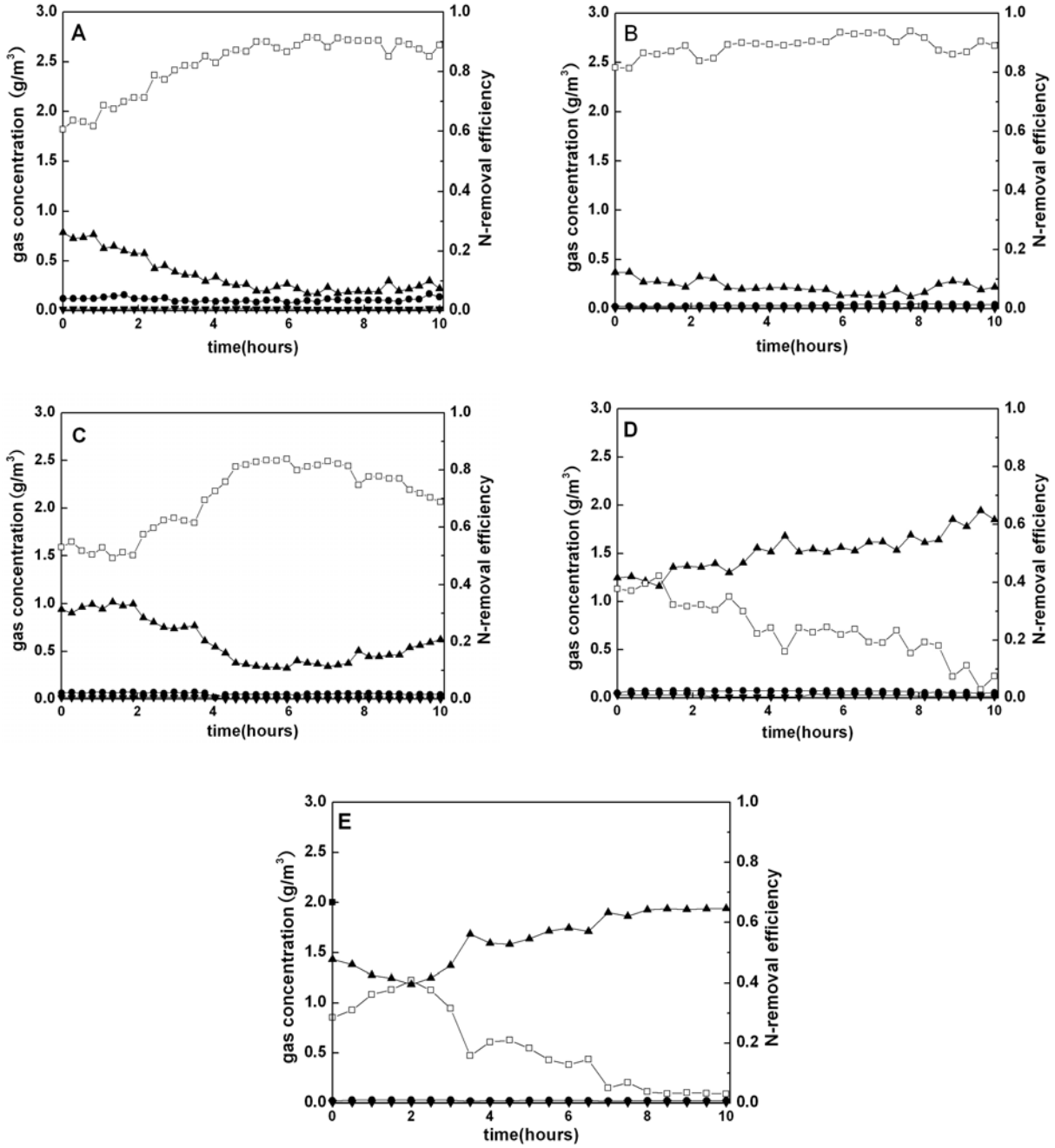


Fig. 3. An entire N-removal (NO+NO_2) cycle. NO feed concentration was kept constant at 2 g/m³. NO removal efficiency (¡%), outlet NO concentration (²%), inlet NO_2 concentration (Ï%) and outlet NO_2 concentration (¼%) are shown as a function of time

So the overall equation is

$$6NO + 2C_3H_6O_3 + 2SO_2 \xrightarrow{coculture}$$

$$3N_2 + 2H_2S + 6CO_2 + 4H_2O$$

Table 3 gives the measured ratios of NO consumed to H_2S produced with the different feed gas compositions. The ratios were in good agreement with the stoichiometric value of 3 according to Eq.4 except the data on the 30th day. One explanation for the negative deviation from stoichiometric value is that part of NO was reduced by heterotrophic denitrifying bacteria using lactic acid as the electron donor (Dasu *et al.*, 1993). It's found that the ratio of Table 3 got closer to the stoichiometric value of 3, indicating that autotrophic denitrification plays a growing prominent role in removing NO with H_2S as the electron donor and so the assumption is reasonable. In the present study, the NO/SO_2 ratio in the feed gas was always

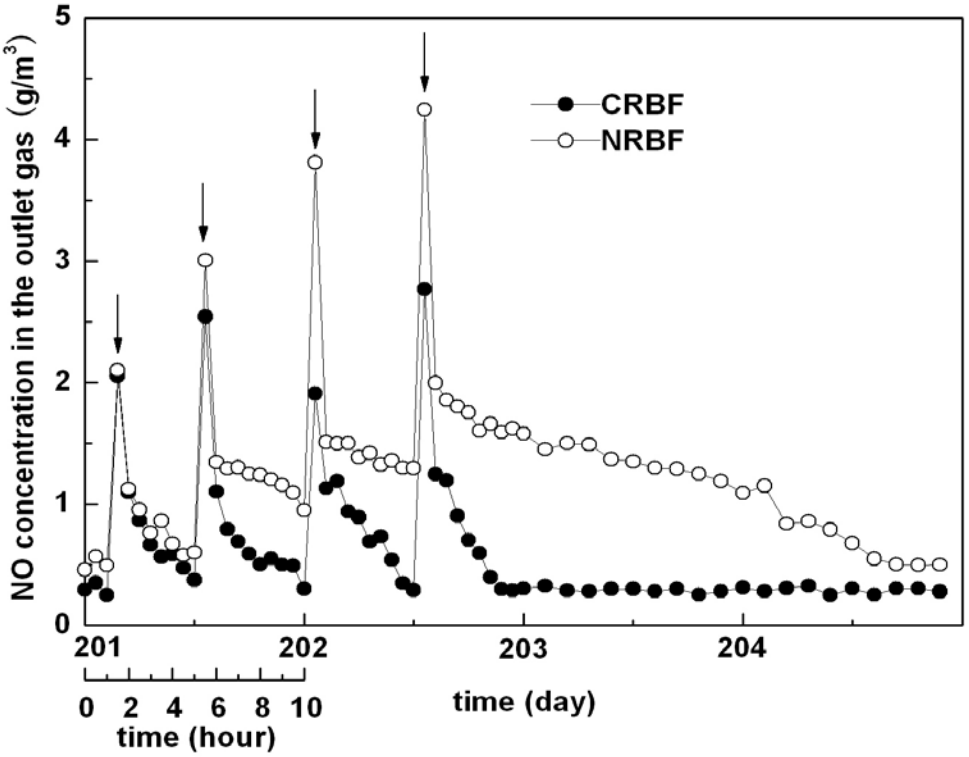


Fig. 4. Comparison of resistance to sudden NO-shock loadings between CRBF and NRBF. NO feed concentration was kept constant at 2 g/m^3 except 1.5th and 5.5th hour of Day 201 and Day 202 at 4.6-4.9 g/m^3 as arrows indicated

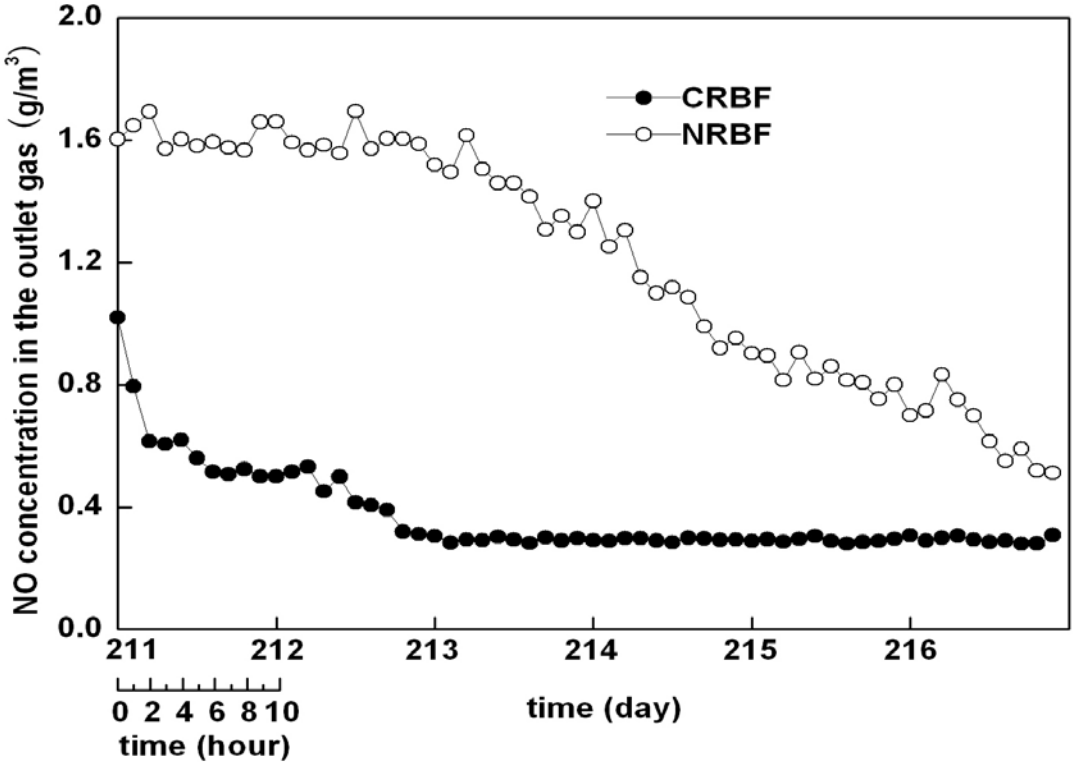


Fig. 5. Comparison of restorability between CRBF and NRBF after 6 days starvation

lower than the stoichiometric ratio of 3 in order to achieve the maximum NO-removal efficiency. Furthermore, the outlet gas of CRBF containing 429-657 mg/m^3 H_2S, together with NO stream, can be introduced into NRBF directly to purify exhaust gas.

CONCLUSION

(1) Approximate 20 days were required for mixed-biofilm development using the landfill leachate as microbial seed in CRBF. FISH-analysis showed that 50% (±15%) of sulfate reducing bacteria and 40% (±15%) of anaerobic denitrifying bacteria were successfully achieved to simultaneously remove NO and SO_2 in CRBF.

(2) When both NO and SO_2 were fed at a constant concentration of 2 g/m^3 with a constant EBRT of 76 s, the SO_2 removal efficiency was always above 95%, while NO removal exhibited an evident periodicity of 5-6 days for the initial 60 days after the attachment phase and then the periodic fluctuation range gradually reduced until a steady-state removal of NO was reached after 130 days with the NO-removal efficiency of around 90%.

(3) Compared with denitrifying bacteria in the NRBF, the coculture in the CRBF had a higher resistance to shock NO-loadings and a better tolerance of starvation.

(4) The stoichiometry of simultaneous bioremoval of NO and SO_2 in CRBF was obtained, which was in excellent agreement with the measured results.

ACKNOWLEDGEMENT

This work was supported by the Specialized Research Fund for the Doctoral Program of Higher Education of China (NO. 20090032110019).

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