

Development of a novel bioscrubbing process for complete treatment of NO_x from flue gases

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ABSTRACT

In this study, a novel bioscrubber was developed for the complete treatment of NO_x from flue gases. As a first step, an autotrophic ANNAMOX system was developed using ammonia as the electron donor and nitrate as electron acceptor generating nitrogen gas as the reaction product. Once an efficient ANNAMOX culture was developed, nitrate was replaced by NO_x. Initially synthetic flue gas with a composition of 80% N₂, 19% CO₂ and 100 ppm_v was fed to the bioscrubber with an EBRT of 60 sec. The system showed a NO_x removal of 20-25 %. The synthetic flue gas with 3-5% oxygen also showed the same NO_x removal efficiency. In the latter case, more ammonia consumption in the system was noted. However, there was no nitrate accumulation in the system in both the cases. In order to improve the NO_x removal efficiency, NO in the flue gas was partially oxidized to NO₂ with the help of ozone (one mole of ozone per mole of NO with an EBRT of 10 sec) and fed to the bioscrubber. The NO_x removal efficiency in the system was improved to 75-80%. The bioscrubber was able to remove more than 90% of the generated NO₃⁻. Performance of the reactor is being monitored at different EBRTs. The new system developed seems to be a promising alternative for the complete treatment of NO_x from flue gases in an environmentally friendly way.

1. INTRODUCTION

The presence of oxides of nitrogen (NO_x) in the ambient air has been, and still is, of great concern because of the toxicity of individual compounds or the secondary pollutants produced by the reaction of NO_x with hydrocarbons and other chemicals such as ozone in presence of sunlight (Wark and Warner, 1981). Nitrogen oxides (NO_x) is a collective name of six compounds namely nitrous oxide (N₂O), nitrogen

dioxide (NO_2), nitrogen trioxide (N_2O_3), nitrogen tetra oxide (N_2O_4) and nitrogen pentoxide (N_2O_5). Among these, NO and NO_2 are of major concerns in air pollution control. From the air pollution point of view, NO and NO_2 are of major concern. NO exists in very low concentrations in the atmosphere and is converted to NO_2 at higher concentrations. About 44% of NO_x pollution is contributed by mobile sources, 55% by stationary sources and the remaining by solid waste disposal and miscellaneous processes. NO_x is responsible for troposphere ozone and urban smog through photochemical reactions. Further, NO_x together with SO_x is the major contributor to the «acid rain» that harms forest crops, buildings, as well as aquatic life.

Since 1970, The U.S. Environmental Protection Agency (EPA) has tracked emissions of the six principal air pollutants – carbon monoxide, lead, nitrogen oxides, particulate matter, sulfur dioxide, and volatile organic compounds. Emissions of all of these pollutants have decreased significantly except for NO_x , which has increased approximately 10%, over this period and nitrogen oxides are facing increasingly stringent regulations due to Clean Air Act Amendments of 1990.

The oxides of nitrogen are formed by the direct combination of oxygen and nitrogen during a variety of thermal processes. Various stationary sources that emit NO_2 are power plants, utility boilers, steel industries, ceramic industry, nitric acid manufacturing industry, oil refineries, ammonia manufacturing industry, fertilizer manufacturers, pickling operations in anodizing plants and nylon intermediate plants. Concern for environmental and health issues coupled with stringent NO_x emission standards indicate a need for the development of efficient low-cost NO_x removal technologies.

Conventional NO_x control technologies include combustion modifications, dry processes and wet processes. The combustion modification technologies such as re-burning, flue gas recirculation, and low NO_x burners, are considered to be suitable for operations with single digit NO_x emission levels (Devahasdin *et al.*, 2003). Dry processes include selective catalytic/non-catalytic reduction (SCR/SNCR) of NO_x to N_2 with ammonia, urea, and hydrocarbons. Wet processes include absorption with liquid phase oxidation, absorption with liquid phase reduction and gas phase oxidation followed by absorption. Among the above mentioned NO_x emission control technologies, the combustion modification (e.g. low NO_x burners) and SCR are the most popular methods (Fujishima *et al.*, 1999).

Combustion modification or pretreatment, a subsidiary of a NO_x treatment system, aims at keeping NO_x production at low. For dry systems such as selective catalytic reduction (SCR) and selective non catalytic reduction (SCNR), elevated temperatures (900-1000°C for SNCR and 270-400°C for SCR) are needed for reasonably good process efficiency. Wet systems have the drawbacks of expensive chemical additives, high water usage, and safety risks from handling of chemicals.

However, most of these control technologies are converting the pollutant from one phase to another or they are highly sensitive and costly. Application of biological process for the flue gas control seems, to be environmentally friendly, economical and sustainable technology.

Biological removal of NO_x from contaminated gas streams is emerging as a novel treatment method. Recently, biofiltration / biotrickling filtration is tried as an attractive alternative for both organic and inorganic air pollutants treatment. It is reported that NO₂ and SO₂ could be removed effectively using biotrickling filter / scrubber (Flanagan *et al.*, 2002; Philip and Deshusses, 2003) due to their high solubility in water. However, not many studies have been carried out on the treatment of acid gases like SO_x and NO_x. Moreover, the tried technologies have many disadvantages like inhibition in presence of oxygen and need high detention times to achieve the necessary degree of treatment. As the solubility of NO in water is very low, biotrickling filter or scrubber is not able to remove NO effectively at a short contact time. Thus, conversion of NO to NO₂ or any other soluble form using suitable technical method and subsequent scrubbing followed by biological denitrification seems to be a viable alternative

Barman and Philip (2005) reported a novel and effective system for the complete treatment of NO_x from flue gases. The system consisted of photocatalytic or ozone oxidation of NO_x, followed by scrubbing and biological denitrification. Maximum photocatalytic oxidation of NO_x was achieved while using powdered TiO₂ at a catalytic loading rate of 10 g/ h, relative humidity of 50%, and a space time of 10 s. The used catalyst was regenerated and reused. A total of 72% of oxidized NO was recovered as HNO₃/HNO₂ in the regeneration process. Stoichiometrically, 10% excess ozone was able to affect 100% oxidation of NO to NO₂. Presence of SO₂ adversely influenced the oxidation of NO by ozone. The scrubbing of NO was effective with distilled water. Heterotrophic denitrifiers were able to denitrify the leachate with an efficiency of 90%, using sewage (COD 450 mg/L) as electron donor. In this system, for the complete treatment, one needs to have three units. It is always advisable to have minimum number of units for the ease in operation and maintenance. Hence, it is essential to try whether the scrubbing and biological denitrification can be achieved in one system.

In this paper, performance of a bioscrubber employing anaerobic ammonia oxidizing bacteria for the removal of NO_x from flue gas was evaluated. Effect of operational parameters such as empty bed contact time, and presence of oxygen on the performance of the system was also studied.

2. MATERIALS AND METHODS

2.1 CHEMICALS

The chemicals used in this study were procured from Ranbaxy Fine Chemicals Limited. A-3, Okhla Industrial Area, Phase-I New Delhi-110 020, India. Nitric Oxide gas was procured from Boruka Gas Limited. Mahadevpua Road, White Field, Bangalore, India.

2.2 SEED SLUDGE

The initial seed sludge was collected from the Sewage treatment plant of IIT Madras Chennai. Cow dung sludge acclimatized for ANNAMOX process in Environmental Engineering Laboratory, Department of Civil Engineering, IIT Madras, Chennai was also used for the study. Settled sludge was added to the reactor (50% of reactor volume, MLSS = 1150 mg/L) for acclimatization. Acclimatization was carried out in twelve separate batch reactors (each having a volume of 100 mL, fitted with air tight septum), under anaerobic environments. During acclimatization, nutrients were added to the reactors for the growth of bacteria. The composition of mineral media is KH_2PO_4 , 0.0572g; NaHCO_3 , 2.1g; NaNO_2 , 0.5915g; $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, 0.3g; $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, 0.2g; FeSO_4 , 0.00625g; EDTA, 0.00625g; $(\text{NH}_4)_2\text{SO}_4$, 0.455g; Trace element solution, 1mL in one liter distilled water.

2.3 ANALYTICAL METHODS

The reactor influent and effluent were analyzed as per Standard Methods (APHA, 1998) in order to monitor the performance of the biological systems. COD of liquid samples was estimated as per standard methods (Ref. No. 5220 Chemical Oxygen Demand, APHA, 1998). Closed reflex method was followed. To estimate the NH_3 -N, NO_2 -N and NO_3 -N concentration, Dionex LC-20 Ion Chromatograph with ED-50 electrochemical detector and IP-25 isocratic pump was used.

2.4 ESTIMATION OF NITRIC OXIDE (NO), NITROGEN DIOXIDE (NO₂)

NO and NO_2 concentration in inlet and outlet gas stream were measured by flue gas analyzer (Quitnox KM 9106). Range NO: 0-5000 PPM, NO_2 : 0- 1000 PPM. Accuracy: +/- 1%. The samples were analysed for the pH, MLSS, MLVSS, and Dissolved Oxygen as per the methods described in APHA, 1998.

2.5 EXPERIMENTAL METHODS

2.5.1 DEVELOPMENT OF ANAMMOX CULTURE FOR NOX REMOVAL

The experiments were carried out in batch systems. A sample volume of 100 mL was taken in a number of serum bottles. Each sample contained 100 mL of minimal medium with 140mg/L of Ammonium-Nitrogen and 84.5 mg/L Nitrate-Nitrogen. The

study was carried out at 25°C in an orbital shaker at 150 rpm under anaerobic environment. Samples were collected at different time intervals and were analyzed for NH₄⁺, NO₃⁻, COD and biomass concentrations. In the second set of experiments, all other conditions were kept the same except the electron acceptor. Instead of NO₃⁻, 152.1 mg/L NO₂⁻-N was supplied to the system.

2.5.2 FED BATCH SYSTEM

The ANAMMOX reactors were operated until the removal of nitrite, nitrate and ammonia reached 80% and the concentrations were brought back to initial concentrations to study the performance of the system in the subsequent removal of nitrite, nitrate and ammonia. Based on the above study, cow dung sludge performed better in presence of nitrite as electron acceptor. Hence, cow dung sludge with nitrite as electron acceptor was used for continuous system.

2.5.3 CONTINUOUS REACTOR STUDIES

The reactor was made out of clear schedule 40 PVC pipe (ID=4cm). The total length of the reactor was 60 cm and bed height was 50 cm. The gas inlet and outlet ports were located at the bottom and top lids of the reactor. The liquid inside the column consisted of mineral medium with the composition specified earlier. There was no external carbon source supplied to the column except for CO₂ and carbonate. The reactor was seeded with about 300 mL of digested cow dung sludge containing a MLSS concentration of 1000 mg/L and remaining 700 mL contained mineral media. During the start-up of the continuous experiment, 150 mg/L of NO₂⁻-N and 230 mg/L of NH₄⁺-N was maintained in the system throughout, since 150-200ppm of NO_x gas was supposed to enter the system daily, with a gas flow rate of 1L/min with a NO_x concentration of 150ppm. Based on the stoichiometry the above concentrations were chosen. The system was operated with a Hydraulic retention time of 24 h. Once the system was stabilized, liquid nitrate feed was replaced by gaseous NO feed.

Simulated flue gas prepared by mixing a metered flow of approximately 1% NO_x (5000 ppm), 10% CO₂, 89% N₂. The gases were homogenized in a mixing chamber and fed to the bioreactor at a flow rate of 1000 mL/min with the help of rotameter. Initial studies were carried out in the absence of oxygen in the system with an EBRT of 60 seconds. The EBRT was gradually reduced upto 10sec. Once, satisfactory performance was observed in the system, 5% of oxygen was introduced in the simulated gas stream and the reactor was operated with a detention time of 60 seconds. The inlet and outlet NO and NO₂ concentrations were measured, ammonia and nitrite concentrations were also measured from the liquid effluent. The mineral media was added to the system and effluent was removed from the system from bottom and allowed to settle in a closed reactor. The supernatant was thrown and the settled biomass

was again added to the system. Further, in order to increase the efficiency of the system, NO was oxidized to NO_2 with the help of ozone.

2.5.4 OXIDATION OF NO BY OZONE

The experiment was conducted in a 5 L borosil bottle at room temperature (27°C). Inlet NO_x concentration was maintained at 150 ppm (NO : 145 ppm and NO_2 : 5 ppm) and space time (10sec) was adjusted by controlling the inlet stream flow rate. Ozone was supplied from ozone generator (Vortex, India) and flow was controlled by controller.

3 RESULTS AND DISCUSSION

3.1 PERFORMANCE OF REACTORS DURING BATCH STUDY

One of the objectives of the study was to check whether NO can be used as an electron acceptor by ANAMMOX bacteria for the generation of di-nitrogen gas. If this route is possible, control of NO_x from flue gases through biological systems can be achieved easily. To check this concept, reactors were developed for the removal of different concentration of nitrite and nitrate in presence of ammonia under anaerobic conditions. Once a strong ANAMMOX culture is developed, it may be easy to replace nitrite and nitrate with NO_x . The performances of these reactors were monitored in terms of COD, nitrite, nitrate and ammonia removal. The MLSS concentrations in the reactors were also monitored periodically.

3.2 PERFORMANCE OF ANAMMOX REACTOR

Ammonia was one of the critical parameters to be monitored to determine the efficiency of ANAMMOX process. Here in this study, ammonia removal in presence of both nitrite and nitrate as electron acceptors were monitored. Two reactors were operated with different seed sludge. Figure 1 a and 1b shows the result of ammonia oxidation in presence of nitrite and nitrate as electron acceptor by both the cultures. Ammonia removal was more in presence of cow dung sludge with nitrite as electron acceptor. This may be due to the prior acclimatization of the sludge for ANAMMOX process. In case of NO_3^- as electron acceptor, both the sludges performed almost in the same manner. The sludge collected from IIT Madras lagoon may be having considerable population of nitrifiers and denitrifiers. The initial concentration of $\text{NH}_4^+\text{-N}$ was 140mg/L and after the test run of 40 days the concentration was reduced to 42 mg/L. $\text{NO}_2^- \text{- N}$ concentration changed from 182 mg/L to 38 mg/L. The stoichiometry of the reaction is as follows



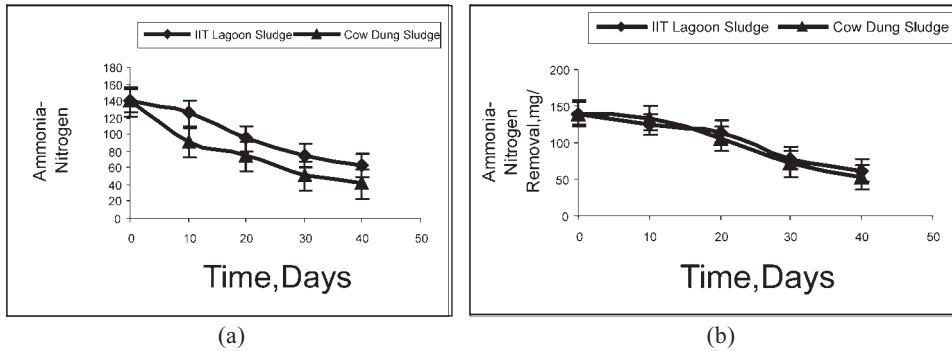


Figure 1. Ammonia oxidation with a) Nitrite b) Nitrate as electron acceptor.

As per reported ANAMMOX Studies (Strous *et al.*, 1998), the ratio of $\text{NH}_4^+ - \text{N} / \text{NO}_2^- - \text{N}$ is 1.3. Stoichiometrically, the reported value and the one obtained in the present study are not matching well. This may be due to the presence of nitrifiers and denitrifiers in more number in the system. A significant quantity of ammonia might be getting oxidized to nitrite/nitrate in the system. It is reported that anoxic nitrification is possible by certain group of microbes under oxygen stress conditions. When nitrate was employed as electron acceptor, the initial concentration of $\text{NH}_4^+ - \text{N}$ was 140 mg/L and after the test run of 40 days the concentration reduced to 52 mg/L. $\text{NO}_3^- - \text{N}$ concentration changed from 88 mg/L to 21 mg/L. The stoichiometry of the reaction is as follows

$$\text{NH}_4^+ - \text{N} / \text{NO}_3^- - \text{N} = 1.31$$

As per reported ANAMMOX Studies (Mulder *et al.*, 1998), the ratio of $\text{NH}_4^+ - \text{N} / \text{NO}_3^- - \text{N}$ is 0.6. Stoichiometrically, the reported value and the one obtained in the present study are not matching well. From this it may be inferred that, the developed sludge has more quantity of denitrifiers. They might have reduced the nitrate to nitrogen gas utilizing the available COD in the system. There was a significant quantity of COD reduction in the system.

The initial MLSS concentration of reactor with cow dung sludge was 1500 mg/L. MLSS concentration gradually increased and after the end of batch study (40 days) the MLSS concentration was 1760 mg/L. Reactor with IIT lagoon sludge was operated with an initial MLSS concentration of 2100 mg/L. At the end of the study, MLSS concentration in the system was 2250 mg/L. Though, the reactor with IIT lagoon sludge had more MLSS, the performance was better in the reactor with cow dung sludge. This must be due to the prior acclimatization of this sludge for ammonia oxidation using nitrite/nitrate as electron acceptors.

3.3 PERFORMANCE OF REACTORS DURING CONTINUOUS STUDY

The batch study clearly showed that cow dung sludge is very effective in the removal of nitrite and ammonia (89% of nitrite and 69% of ammonia) and hence, cow dung sludge was used for the continuous studies. The system was run continuously on a sequential batch mode with a detention time of 24 h and with initial concentration of 150mg/L of NO_2^- . Initially the study was carried out without introducing NO_x gas. The significance of this study was to make the system get acclimatized to nitrate removal with ammonia oxidation. The amount of nitrate was chosen in such a way that, that will be the total nitrite/nitrate generated in the system by the scrubbing of NO_x from flue gas for a flow rate of 1L/min with a concentration of 100 ppm_v during 24 h. The removal of nitrite and ammonia soon after 24 h was monitored. Every 24 h interval, the concentration of nitrate and ammonia was made up to the initial concentration.

3.4 AMMONIA REMOVAL WITH NITRITE AS ELECTRON ACCEPTOR IN THE CONTINUOUS SYSTEM

The study of ammonia oxidation with nitrite as electron acceptor was carried out in batch systems with a longer hydraulic retention time. To assess the ability of developed ANAMOX culture to remove nitrate, a continuous system was operated with a high loading. The results are presented in Figures 2a and 2b.

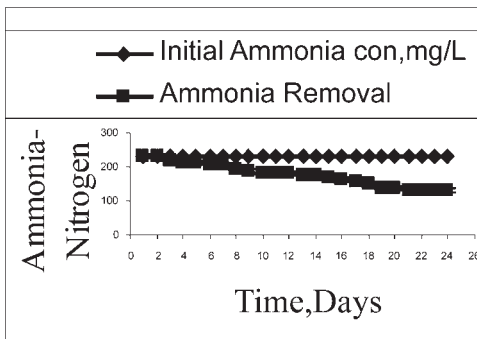


Figure 2a. Ammonia removal in the system.

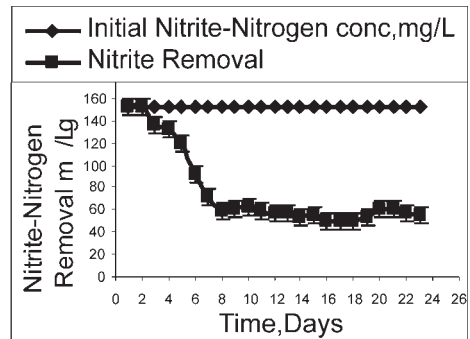


Figure 2b. Nitrite removal in the system.

After 25 days of acclimatization, the continuous system showed 71% of ammonia removal and 76% of nitrite removal with a detention time of 24 h. The initial concentration of $\text{NH}_4^+\text{-N}$ was 233 mg/L and after 23 days of continuous operation the concentration reduced to 131.4 mg/L. The corresponding nitrite removal was from 152 mg/L to 35 mg/L. The stoichiometry is as follows $\text{NH}_4^+\text{-N} / \text{NO}_2^-\text{-N} = 0.87$. Hence from the above study, it can be inferred that ammonia oxidation and nitrite removal is due to the presence of nitrifiers and denitrifiers.

3.5 PERFORMANCE OF REACTORS DURING CONTINUOUS STUDY AFTER INTRODUCING NO_x GAS

3.5.1 STUDY IN ABSENCE OF OXYGEN

Initially the study was carried out in the absence of oxygen. During the entire study period, inlet NO_x concentration of 100 ppm was maintained in the system and the outlet concentration was measured regularly. The results of the study is presented in Fig 3 a. After the system was operated continuously for 11days, the outlet NO_x concentration was reduced to 78 ppm. The above results indicate that, the system performance in NO_x removal was very less with a percent NO_x gas removal of 22%. This may be due to the very low solubility of NO_x gas.

3.5.2 STUDY IN PRESENCE OF OXYGEN

Flue gas always has 3-8% of oxygen. The performance of the system was evaluated in presence of 5% oxygen. Ammonium gets oxidized to nitrite in presence of oxygen with the help of autotrophic nitrifiers and ammonium reacts with nitrite to produce nitrogen gas. This is the possible mechanism in presence of oxygen. Study was carried out with an inlet NO_x concentration of 100 ppm along with 5% oxygen keeping all other parameters the same. Outlet NO_x concentration was monitored regularly. After 11 days the continuous operation, the outlet NO_x concentration was 80ppm and the percentage removal of NO_x was 20% (Figure 3b). Though presence of oxygen did not hinder the activity of the biological system, the overall removal efficiency was very low. The low solubility of NO_x must be the reason for this.

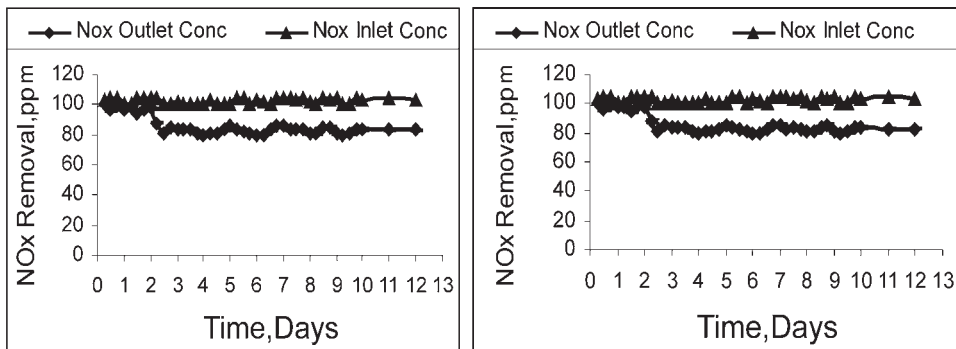


Figure 3a. NO_x removal in absence of oxygen. Figure 3b. NO_x removal in presence of oxygen.

3.6 PERFORMANCE OF REACTORS DURING CONTINUOUS STUDY AFTER OZONE OXIDATION

From earlier study results, it is clear that, using the new biosystem, it is not possible to remove NO_x in the form of NO due to its low solubility. Hence, in order to increase the efficiency of NO_x removal, first NO was oxidized with the help of ozone and allowed the NO₂ thus formed to enter in the biosystem. Inlet and outlet NO and NO₂ concentrations were measured along with nitrite and ammonia removal in the aqueous phase of the biosystem. The optimization of NO oxidation by ozone was done by Barman and Philip (2005). These results were used for the present study. A NO_x concentration of 200 ppm (NO: 175 ppm and NO₂: 25 ppm) was used. Ozone flow was gradually increased and the oxidation of NO to NO₂ was monitored. It was found that almost all NO was converted (99%) to NO₂ when slightly excess (10%) of stoichiometric amount of ozone was passed.

3.7 NO_x REMOVAL AFTER OZONE OXIDATION

NO_x removal studies were carried out with an inlet NO_x concentration of 150 ppm (NO₂=145 ppm; NO =5 ppm) and with an EBRT of 60 sec. Gradually the contact time was reduced to 30 sec and then to 10sec. The results are presented in Figure 4 a. The results clearly indicate that the efficiency of NO_x removal has increased significantly after the partial oxidation of NO to NO₂. After 15 days of operation, a NO_x removal efficiency of 79% was achieved in the bio-system. In order to understand the system behavior at varying detention times, the study was carried out with 30sec and 10sec. when the detention time was reduced to 30 sec., the efficiency of the system was reduced to 60%. Further reduction in EBRT to 10 sec. deteriorated the performance of the system. This must be due to the less contact of the pollutant with the bio-scrubbing medium. The addition of packing material or reducing the bubble size by modifying the diffuser marginally improved the efficiency of the system.

3.8 NITRATE REMOVAL IN THE SYSTEM

Fig 4b shows the nitrate removal from the system after ozone oxidation. Initially the nitrate concentration in the system was increased during the study. But after 3 days, nitrate concentration gradually came down and reached a steady state in 12 days.. After reaching a steady state nitrite removal of 75%, there was no further improvement. The residual nitrate concentration may be the result of ammonia oxidation. However, as the nitrate loading to the system was increased (by reducing the EBRT and increasing the gas flow rate) the residual nitrate concentration at steady state also was increased marginally. At 30 Sec retention time, 55% of nitrate removal was observed where as 10sec retention time (gas) gave a nitrate removal efficiency of 45%.

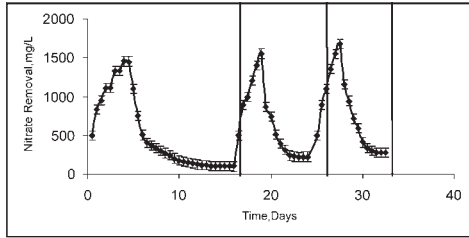
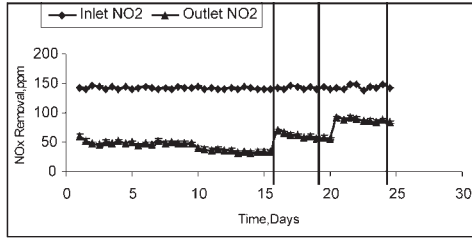


Figure 4b. Nitrate removal in the system.

Figure 4a. NO_x removal in the continuous system after ozone oxidation.

3.9 NO_x REMOVAL IN THE CONTROL REACTOR

The system was operated at a detention time of 60 sec. The results are presented in Figure 5a. There was about 74% NO_x removal initially and gradually the NO_x removal efficiency was reduced to 68%. From this result, it is clear that, the major mechanism of NO₂ removal is scrubbing. When microbes are present in the system, the accumulated nitrate is utilized. This gives a favorable concentration gradient for NO_x removal. This may be the reason for a better performance while microbes are present in the system.

3.10 NITRATE REMOVAL IN THE CONTROL REACTOR

Figure 5 b shows the abiotic nitrate removal from the system. As the time progressed, the nitrite concentration was kept on increasing. This shows the absence of autotrophic ANAMMOX culture, which was earlier responsible for the nitrite removal from the system 150 ppm of NO₂ gas was allowed to enter the system for a detention time of 1 min and this was getting converted to nitrate in the system. In presence of autotrophic culture the system was responsible for removal of nitrate to 75% but in the absence of microbes nitrate was accumulated in the system.

4. CONCLUSIONS

The following conclusions can be made based on the investigation carried out in the present study. Batch experiments indicated that, ammonia oxidation and simultaneous nitrite removal is possible in presence of cow dung sludge. Maximum removal efficiency of 79% of NO_x gas at detention time of 60 sec, 60.1% at 30 sec, 40.33% at 10 sec was achieved after the ozone oxidation. Less removal of NO_x was observed before ozone oxidation due to the low solubility of NO gas. The biological system containing ANAMMOX cultures seems to be a feasible option of treating nitrogen oxides from flue gases.

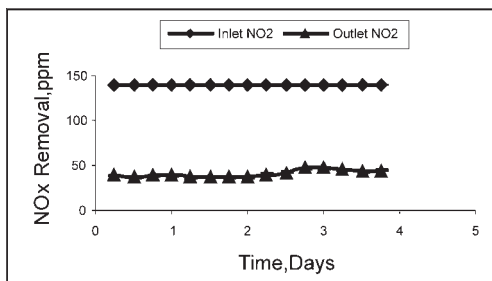


Fig. 5a. NOx removal from the system (Blank).

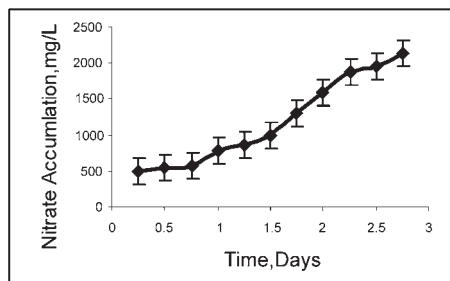


Fig. 5b. Nitrate accumulation in the system (Blank).

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