

**SELECTIVE NON-CATALYTIC REMOVAL (SNCR) OF
NITROGEN DIOXIDES (NO₂) USING AMMONIA**

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for presentation and publication at the

2nd Joint Meeting of the United States Sections of the Combustion Institute
2001 Spring Technical Conference

The Oakland Marriott City Center
Oakland, CA

25 – 28 March 2001

11 March 2001

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The selective non-catalytic removal (SNCR) process using ammonia was examined for the removal of nitrogen dioxide (NO₂). In the past, SNCR processes have focussed primarily on the species nitric oxide (NO). For most combustion processes, NO₂ is often less than 10% of the total nitrogen oxides (NO_x). Nevertheless, even these small amounts are important as the regulations concerning NO_x continue to become stricter. Furthermore, for a complete understanding of the exhaust gas nitrogen chemistry, the reactions relating to both NO and NO₂ are important.

The exhaust gas was simulated by using bottled gases to insure accurate and steady species concentrations. These gases flowed through an electrically heated, laminar flow reactor for gas temperatures between 800 and 1300 K. A range of simulated exhaust gas was used containing various concentrations of oxygen and carbon monoxide. A Fourier transform infrared (FTIR) spectrometer was used to determine concentrations of all species. NO₂ removal was a strong function of temperature and gas composition. NO₂ removal was much greater for the cases using ammonia especially for temperatures less than about 1100 K; and NO₂ removal was greater for the lower oxygen cases. Carbon monoxide enhanced the removal of NO₂ for the cases with no ammonia.

INTRODUCTION

The emission of pollutant gases from combustion devices continues to be a current concern, and will be of even higher priority in the future [1–3]. For today's industrial and private applications of combustion devices such as boilers or internal combustion (IC) engines, toxic and harmful gases must be minimized to acceptable levels. Such unwanted products of combustion processes are species such as CO, NO, NO₂, N₂O and others. As a result, the control of nitric oxides (NO_x = NO + NO₂) is important for many processes today.

Nitric oxide (NO) and nitrogen dioxide (NO₂) are two of the major pollutant gases produced by combustion processes involving exothermic reactions with air. Because most combustion processes use air as the oxidant, nitrogen is always present. Due to the high temperatures of combustion processes and with nitrogen and oxygen available, nitric oxides are produced [1].

A number of NO_x removal processes are available [1–3]. One such technique is selective non-catalytic removal (SNCR) which is often used in modern post combustion treatment of exhaust gases. SNCR processes use a reducing agent like ammonia (NH₃) which is injected into the exhaust stream. For optimal injection processes, all of the gases (including NO and NO₂) in the exhaust stream are exposed to the reducing agent. For appropriate conditions and following a series of reactions, the nitric oxides are converted to N₂ [1, 3].

Nitrogen dioxide is directly formed in combustion processes or results from reactions of NO with oxygen. For most combustion processes, NO₂ is typically less than 10% of the total NO_x [1]. In spite of this relatively small fraction, the emitted NO₂ is a concern. Gaseous NO₂ is more toxic than NO, and has a brown color whereas NO is colorless. Other specific concerns include that NO₂ contributes to the greenhouse effect (the

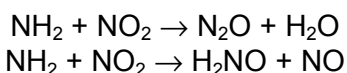
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Proceedings of the 2001 Technical Meeting of the Joint US Sections of the Combustion Institute.

proposed global warming process), and photochemical smog. Another effect of NO₂ on the environment is in the formation of acid rain which occurs in combination with other chemicals [2]. Furthermore, for a complete understanding of the exhaust gas nitrogen chemistry, the reactions relating to both NO and NO₂ are important.

Previous Work: In general, studies of NO₂ removal by SNCR processes have been rather limited. One study which did address this topic was reported by Glarborg *et al.* [4] in 1995. Their experiments included the temperature range between 850 and 1350 K as well as gas compositions that are representative of exhaust gases. Inlet concentrations that they used included: 547 ppm NH₃; 285 ppm NO₂; 35 ppm NO; trace amounts of H₂O and O₂; zero CO; and balance N₂. For their study, the major products of the experiments were NO (about 340 ppm at 1150 K) and N₂O (about 15 ppm at 1150 K). Although this study was for trace oxygen and did not include CO, the results provided insight on the important reactions involving NO₂.

Glarborg *et al.* [4] found, for the conditions of their experiment, that NO₂ conversion follows two channels:



Their results suggest that the first reaction is important at the lower temperatures and the second reaction is important at the higher temperatures.

A number of related investigations have focussed on the conversion of NO to NO₂. An example of this type of study was completed by Hunderup and Roby [5]. They used a high pressure (up to 10 atm) flow reactor to study the conversion of NO to NO₂ in a gas stream similar to that of a gas turbine combustor exit. For their conditions, they showed significant conversion only for temperatures between approximately 800 and 1000 K.

Another category of work regarding NO₂ is more relevant for temperatures typical of combustion. For example, He *et al.* [6] investigated the reactions between HNCO and NO₂ for temperatures from 1700 to 2500 K in shock waves. This study supports the fact that NO₂ may be converted to NO, but the high temperature range and the fact that shock waves were investigated may preclude the application of these results to NO₂ removal in SNCR processes.

The current work, therefore, is directed to further the understanding of NO₂ chemistry in

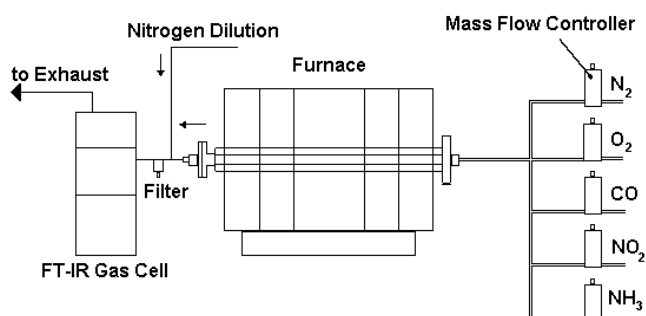


Figure 1. Schematic of the experimental apparatus.

SNCR processes. Specifically, this work examined the removal of NO₂ for an SNCR process using ammonia in gas streams with different oxygen and carbon monoxide concentrations.

DESCRIPTION OF THE EXPERIMENTS

In the present study, simulated exhaust gas was used to conduct a flow reactor study on the selective non-catalytic removal (SCNR) of nitrogen dioxide using ammonia as the reducing agent. For this purpose the gas components of the simulated exhaust gas (NO₂, CO, O₂, N₂) were mixed with ammonia prior to the reaction zone. Simulated exhaust gas has been shown to be an acceptable substitute for real exhaust gas by Siebers and Caton [3].

Figure 1 is a schematic of the experimental equipment. The gases were stored in standard gas cylinders under pressures up to about 100 bar. Prior to the entrance to the mass flow controllers, the pressure was regulated to about 4 bar. After the flow controllers, the desired composition was mixed at about atmospheric pressure. Teflon tubes were used to transport the gas to the reactor entrance. This region was subject to tape heating to ensure preheating of the gas mixture above the vaporization point of water. The gas mixture passed through a section with increasing diameter and small baffles, then the gases entered the 3-stage furnace.

The reactor was a straight steel pipe which was lined with a quartz tube (ID of 1.8 cm) to minimize any catalytic surface reactions. The flow in the reactor was laminar with Reynolds numbers below 100. The total flow through the reactor was 1100 sccm¹, and was the same for all experiments. The residence time in the reactor was estimated to be between 1.3 sec (at 1300 K) and 2.1 sec (at 800 K). The reactor quartz tube was

¹ The standard conditions were 0°C and 1 atm where "sccm" is "standard cubic centimeters per minute."

Table 1. Experimental Conditions for the Nitrogen Dioxide Experiments

Parameter	Value
Temperature Range	800 – 1300 K
Residence Time	1.3 – 2.1 sec
Residence Time Expression (secs)	$= 1705/T$ (K)
Total Reactor Flow	1100 sccm
Inlet Species:	
Nitric Oxide (NO)	0 ppm
Nitrogen Dioxide (NO ₂)	420 ppm
Ammonia (NH ₃)	0, 760 ppm
Oxygen (O ₂)	0.73; 13.0%
Carbon Monoxide (CO)	0, 100, 600 and 900 ppm
Nitrogen (N ₂)	Balance

sealed from the outer steel tube by using Grafoil sealing tape. This prevented gas from flowing between the quartz and steel tube, and hence, prevented any reactions due to the steel surfaces.

After passing through the furnace, the gases were diluted by 5000 sccm of nitrogen to prevent further reactions and to decrease the temperature to a level consistent with use in the gas analyzer. The excess nitrogen also prevented the water vapor in the mixture from condensing which could have affected the chemical composition of the gases. The gases passed through a 0.5 micron filter, and then entered the gas cell before being exhausted out of the laboratory.

The product gases from the reactor were analyzed with a Fourier transform infrared (FTIR) spectrometer. The FTIR spectrometer is manufactured by Bio-Rad, model FTS 60A, and possesses dynamic alignment with up to 0.1 cm⁻¹ resolution. The FTIR is ideally suited to this application due to its ability to provide on-line analysis of a wide variety of species.

The FTIR was calibrated with known concentrations of the relevant species. The specific wave numbers used for each species are listed in appendix A. The calibrations were indirectly verified by comparison with species balances of certain atoms (e.g., nitrogen and carbon atoms). Some discrepancy (less than about 20%) was noted for the higher concentrations of carbon dioxide. Since the absolute values of carbon dioxide were not needed, this discrepancy was not a concern.

Experimental Conditions: Table 1 is a summary of the major experimental conditions for this

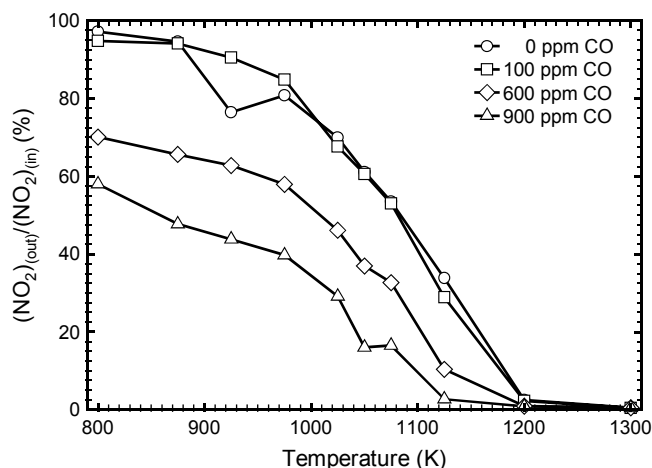


Figure 2. NO₂ conversion as a function of temperature for four carbon monoxide concentrations for initial conditions of 420 ppm NO₂, 0 ppm NH₃, and 0.73% O₂.

study. Over the course of all of the experiments, the average nitrogen dioxide concentration was about 420 ppm. For a given experiment, however, the actual concentration used was 420 ± 40 ppm, (but was constant during the specific experiment). Further details of the experiment are available from Gentemann [7].

RESULTS

Three sets of experiments (each with specific inlet concentrations) were conducted. The first set was conducted with no ammonia. This case, therefore, provided data so that the influence of the temperature on the NO₂ chemistry could be determined without the presence of ammonia. The second set of experiments was with the use of ammonia (760 ppm) with low oxygen concentration (0.73%). This results in a ratio² of ammonia to inlet NO₂ of about 1.8. The third set of experiments was with the same ammonia concentration, but with 13% oxygen. For each set of experiments, carbon monoxide was varied up to 900 ppm.

Experiments with no Ammonia: Figure 2 shows the ratio of (NO₂)_{out} and (NO₂)_{in} as a function of temperature for four carbon monoxide concentrations for no ammonia and 0.73% oxygen³. In general, the conversion of NO₂ increased (i.e., the ratio of (NO₂)_{out} and (NO₂)_{in} decreased) as temperature increased. This means that the NO₂ was converted to other nitrogen species (e.g., NO, N₂O, N₂, ...). The conversion of NO₂ is

² In the literature, this ratio is often referred to as the β -ratio.

³ This amount of oxygen, 0.73%, was the minimum possible since the NO₂ was required to be stored with balance air to insure stability.

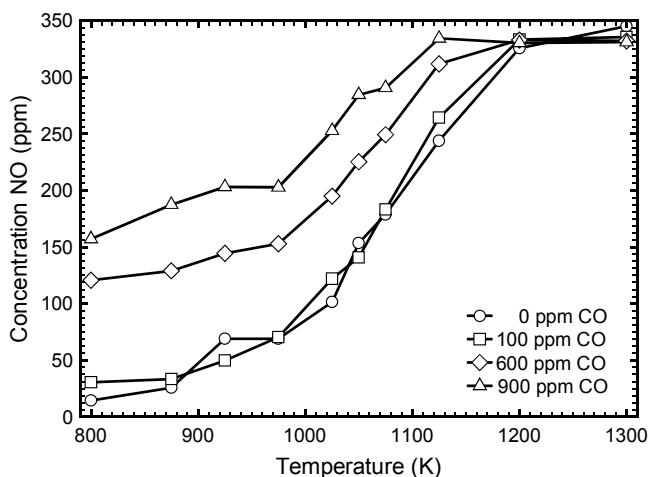


Figure 3. Nitric oxide concentration as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 0 ppm NH_3 , and 0.73% O_2 .

higher for higher carbon monoxide concentrations for each temperature.

Figure 2 shows that the NO_2 conversion is on the order of 5% for 0 and 100 ppm CO at 800 K. This NO_2 conversion continues to increase to almost 100% at 1200 K. The cases for 600 and 900 ppm CO show similar NO_2 conversion with respect to temperature, but resulted in lower NO_2 concentrations. For 800 K, for the cases of 600 and 900 ppm CO about 30% and 42% conversion, respectively, was observed.

Figure 3 shows the NO concentration as a function of temperature for these conditions. In general, the NO concentration increased as temperature increased. For 0 and 100 ppm CO, about 25 ppm of NO was produced at 800 K. The amount of NO increased up to a level of about 330 ppm for 1200 and 1300 K. For the cases of 600 and 900 ppm CO, about the same amounts of NO were produced at 1200 and 1300 K. In general, for temperatures less than about 1200 K, the concentration of NO was higher for the cases with higher concentrations of CO. For example, at 800 K, the nitric oxide concentration was about 125 ppm for 600 ppm CO and was about 160 ppm for 900 ppm CO.

Figure 4 shows the normalized total nitrogen atom balance (including NO, NO_2 and N_2O^4) as a function of temperature. Numbers less than 100% indicate the probable formation of molecular nitrogen (which could not be detected by the FTIR). In general, as the temperature increased,

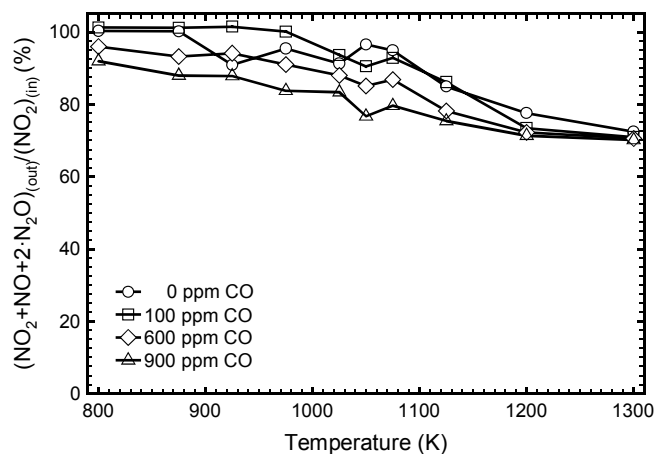


Figure 4. Detectable nitrogen atom balance as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 0 ppm NH_3 , and 0.73% O_2 .

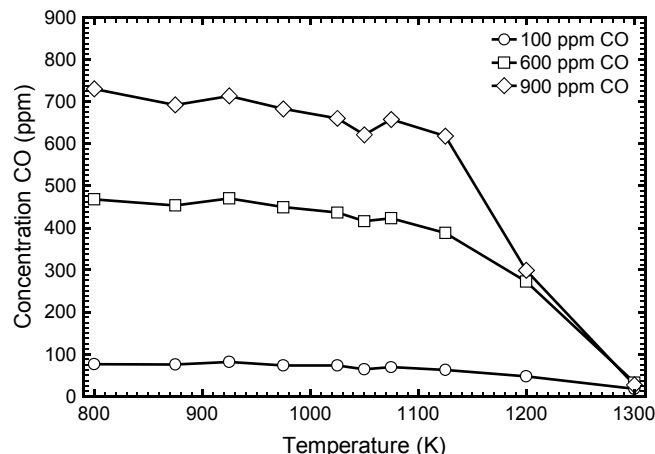


Figure 5. CO concentration as a function of temperature for three carbon monoxide concentrations for initial conditions of 420 ppm NO_2 , 0 ppm NH_3 , and 0.73% O_2 .

the conversion to molecular nitrogen increased. For a temperature of 800 K, the net removal of NO_2 (the amount of NO_2 that reacts to molecular nitrogen) was between about 0% (for 0 and 100 ppm CO) and about 9% (for 900 ppm CO). For higher CO concentrations, the conversion to molecular nitrogen increased. For temperatures above about 1100 K, the CO concentration became less important. The conversion approached a value of about 30% at 1300 K for all carbon monoxide cases.

Figure 5 shows the CO concentration as a function of temperature. The CO concentration decreased slightly for all cases up to about 1125 K. For higher temperatures, the high CO concentration cases (600 and 900 ppm CO) showed a strong decrease down to about 25 ppm for all cases at 1300 K. The removed CO was

⁴ For the cases with no ammonia, N_2O production was always less than about 4 ppm.

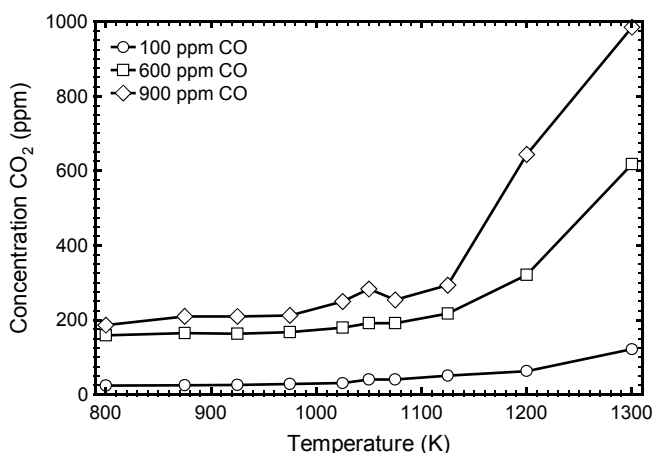


Figure 6. CO₂ concentration as a function of temperature for three concentrations of carbon monoxide for initial conditions of 420 ppm NO₂, 0 ppm NH₃, and 0.73% O₂.

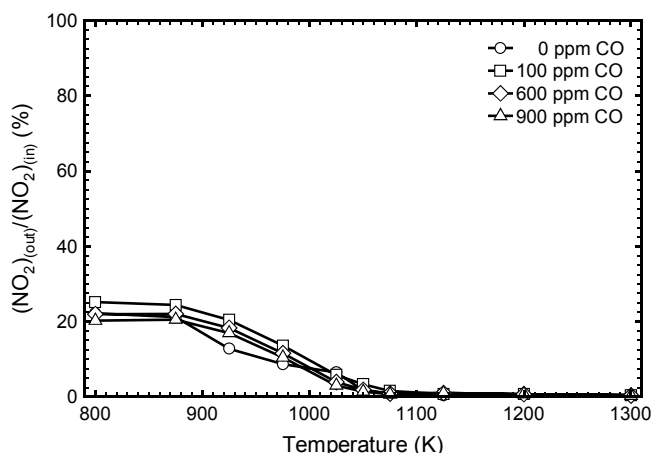


Figure 7. NO₂ conversion as a function of temperature for four carbon monoxide concentrations for initial conditions of 420 ppm NO₂, 760 ppm NH₃, and 0.73% O₂.

converted to CO₂ which can be observed in figure 6. An increase in CO₂ concentration was reported for all cases over the whole temperature range. For temperatures below 1125 K, CO₂ production was modest. Above 1125 K, however, a more rapid increase in the CO₂ concentration was observed which is consistent with the decrease in CO concentration reported in figure 5.

Cases with Ammonia (low oxygen): Figure 7 shows the normalized NO₂ concentration as a function of temperature for the case of 0.73% oxygen and 760 ppm of ammonia. Significant NO₂ conversion was observed even at the lowest temperature of 800 K – on the order of about 80% and this conversion was slightly enhanced by increasing CO concentrations. This tendency could be observed over the whole temperature range. Increasing temperature lead to a further

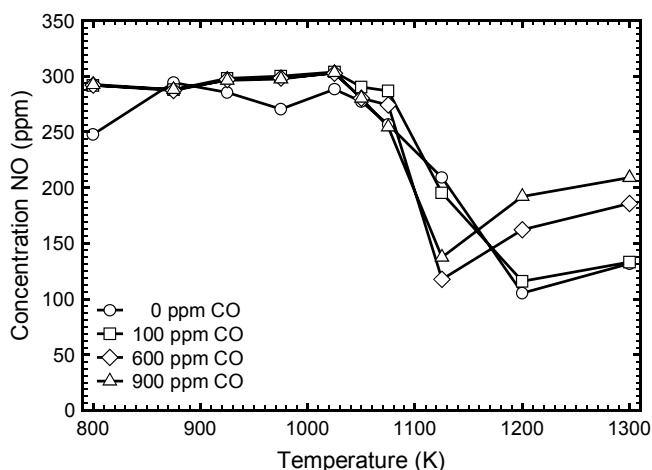


Figure 8. NO concentration as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO₂, 760 ppm NH₃, and 0.73% O₂.

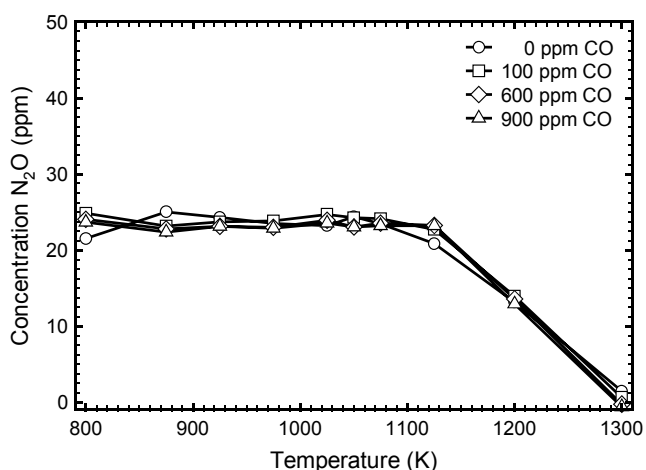


Figure 9. N₂O concentration as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO₂, 760 ppm NH₃, and 0.73% O₂.

decreased NO₂ concentration up to almost complete conversion at temperatures above 1075 K.

The converted nitrogen dioxide reacted at least partially to nitric oxide as shown in figure 8. For temperatures below about 1000 K, the nitric oxide concentration was roughly constant at about 300 ppm. In general, for higher temperatures, the nitric oxide concentration decreased. The effect of carbon monoxide on NO production for the higher temperatures was mixed.

For this case, figure 9 shows nitrous oxide (N₂O) concentration as a function of temperature. Between about 800 and 1125 K, the N₂O concentration was observed to be about constant with a value on the order of 25 ppm. For higher temperatures, N₂O concentrations decreased down to concentrations of about zero for 1300 K.

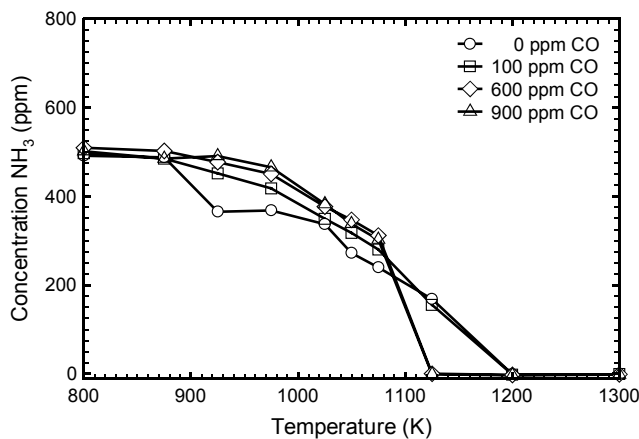


Figure 10. Ammonia concentration as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 0.73% O_2 .

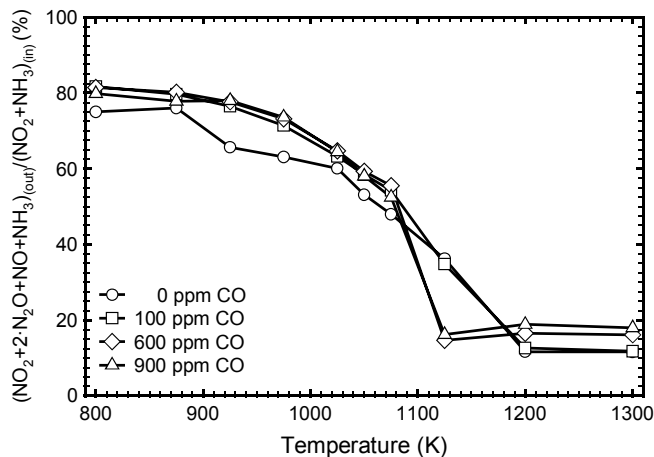


Figure 11. Detectable nitrogen atom balance as a function of temperature for four concentrations of carbon monoxide for inlet conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 0.73% O_2 .

Figure 10 shows the concentration of ammonia emitted from the process (known as ammonia slip) as a function of temperature and oxygen level. In general, the ammonia decreased as the temperature increased, and was not a strong function of the CO concentration. This led to the ammonia slip approaching zero for high temperatures (>1100 K).

Figure 11 shows the normalized nitrogen atom balance as a function of temperature for these cases. In general, removal of detectable nitrogen containing species was not a strong function of CO concentration. For 800 K, removal of 20% was observed, and this removal increased with increasing temperature. Above about 1200 K, the removal reached values between 80 and 90% for all cases.

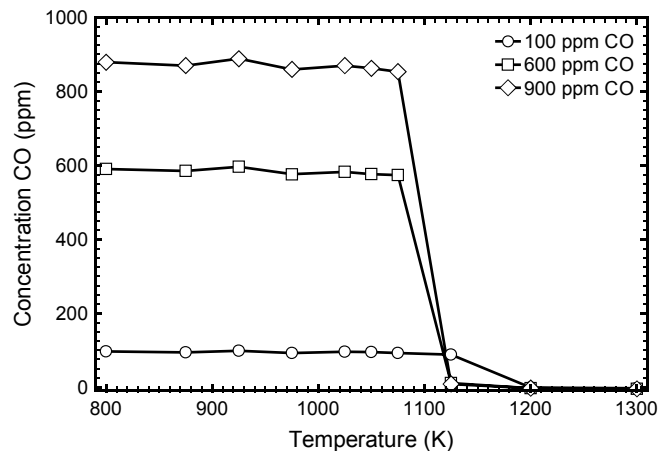


Figure 12. CO concentration as a function of temperature for three concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 0.73% O_2 .

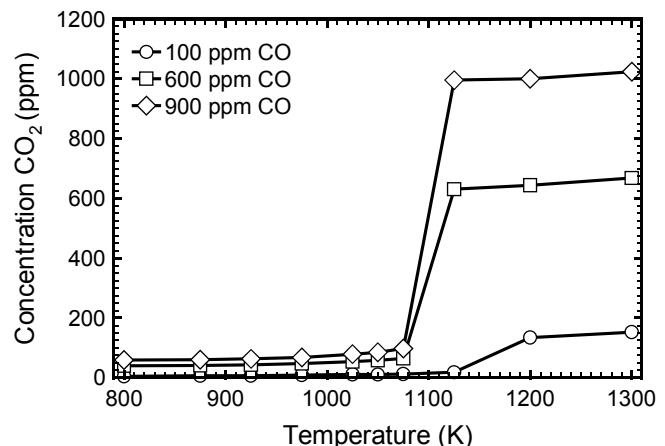


Figure 13. CO_2 concentration as a function of temperature for three CO concentrations for inlet conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 0.73% O_2 .

As before, the carbon monoxide (CO) and carbon dioxide (CO_2) concentrations (figures 12 and 13, respectively) show the expected opposite behavior. For temperatures between 800 and 1075 K, constant levels of CO were observed, whereas for higher temperatures, near complete CO removal was found. Figure 13 shows the CO_2 concentration as a function of temperature. Very low CO_2 production was found for temperatures between 800 and 1075 K. For higher temperatures, increased production of CO_2 was observed. These results can be contrasted to the results with no ammonia (figures 5 and 6). The influence of the ammonia is to enhance the CO oxidation at temperatures of greater than about 1100 K.

Cases with Ammonia (high oxygen): Figure 14 shows the normalized NO_2 concentration as a function of temperature for the case of 13.0%

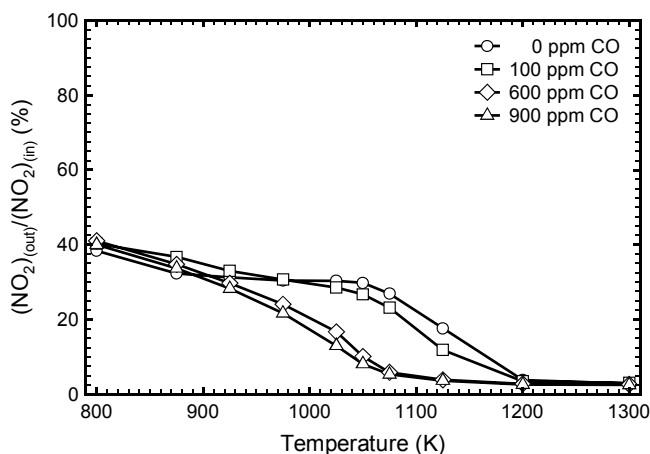


Figure 14. NO_2 conversion as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 13.0% O_2 .

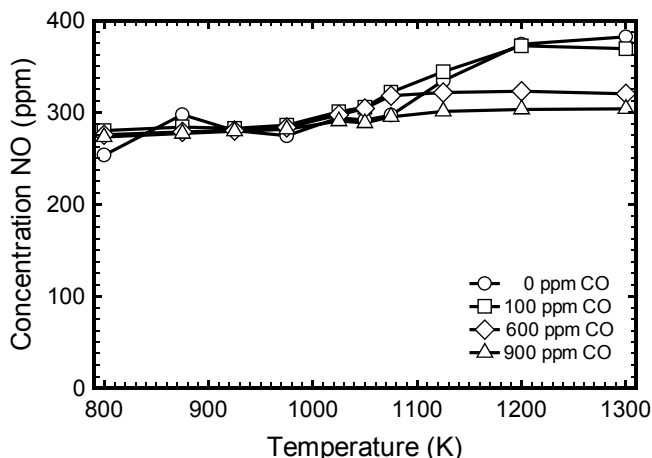


Figure 15. NO Concentration as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 13.0% O_2 .

oxygen. From an initial NO_2 removal of about 60% for all cases of CO concentration, a decrease of NO_2 concentration with increasing temperature was observed. The CO concentration had a weak influence: for temperatures below 1200 K, higher CO concentration showed a slightly higher NO_2 conversion at lower temperatures. This tendency was not observed for the temperatures between about 1200 and 1300 K, where a constant NO_2 removal of 95% was achieved.

Figure 15 shows the NO concentration as a function of temperature for these cases. From a concentration of about 280 ppm at 800 K, NO increased with increasing temperature. Above a temperature of 1025 K, lower concentrations of CO lead to higher NO concentrations. These results for high oxygen can be compared to the re-

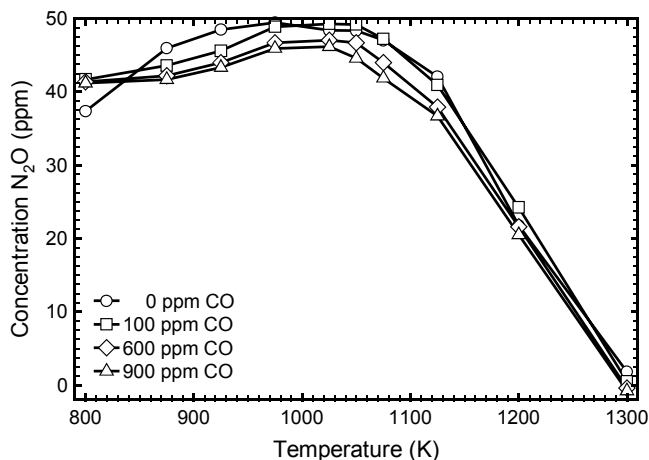


Figure 16. N_2O concentration as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 13.0% O_2 .

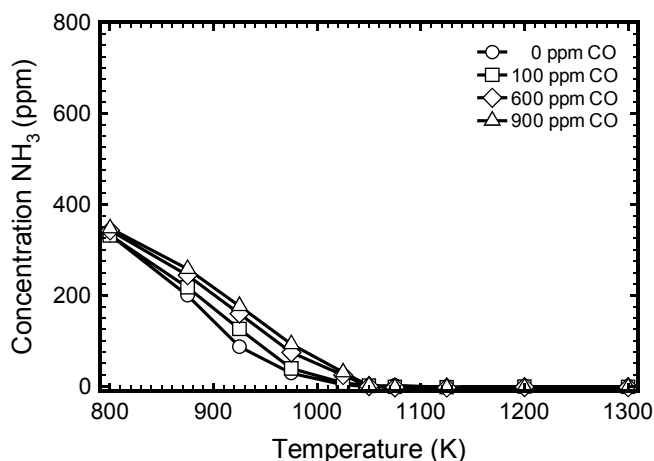


Figure 17. Ammonia concentration as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 13.0% O_2 .

sults for low oxygen in figure 8. This comparison leads to the conclusion that conversion of NO_2 to NO is enhanced with temperature for higher oxygen cases.

For this case, figure 16 shows nitrous oxide concentration as a function of temperature. As shown, the N_2O concentration was about 42 ppm at 800 K for all carbon monoxide cases. Up to a temperature of 1050 K, the N_2O concentration slightly increased, and this increase was enhanced for lower CO concentrations. Above that temperature (1050 K), the nitrous oxide concentration decreased to about zero for 1300 K for all cases.

Ammonia slip (figure 17) was found to be on the order of 350 ppm at 800 K for all cases. For increasing temperature, the ammonia exit concentration decreased. For temperatures greater

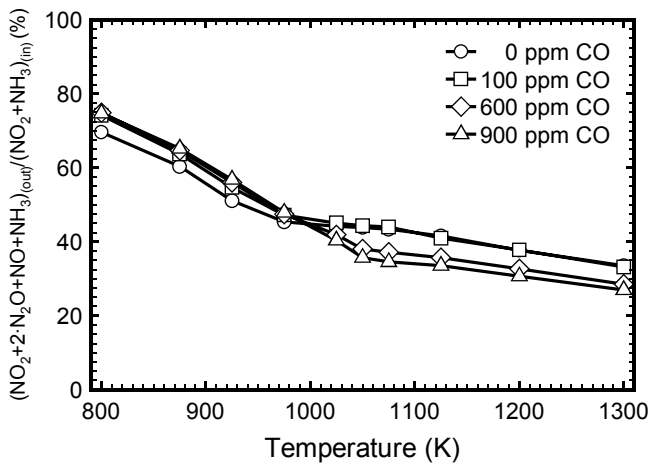


Figure 18. Detectable nitrogen atom balance as a function of temperature for four concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 13.0% O_2 .

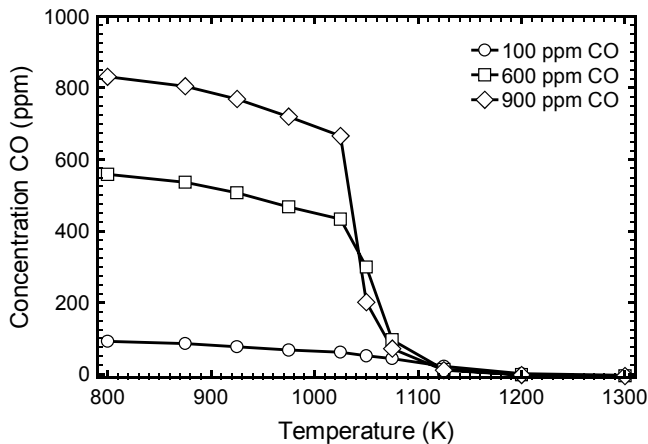


Figure 19. Exit CO concentration as a function of temperature for three inlet concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 13.0% O_2 .

than about 1050 K, no significant amount of NH_3 could be found for any CO concentration case. These results can be compared to figure 10 for the lower oxygen case. As shown, the higher oxygen case promotes the removal of ammonia for the same conditions.

Figure 18 shows the normalized nitrogen atom balance as a function of temperature for the case of 13.0% oxygen. In general, the "N" containing species continually decrease for increasing temperature. For the higher temperatures, higher CO concentrations lead to slightly higher removal characteristics, which resulted in a maximum removal of about 70% for the case of 900 ppm CO and a temperature of 1300 K. In comparison with the lower oxygen case (figure 11), for the higher temperatures (say, above about 1100 K), the net conversion to molecular

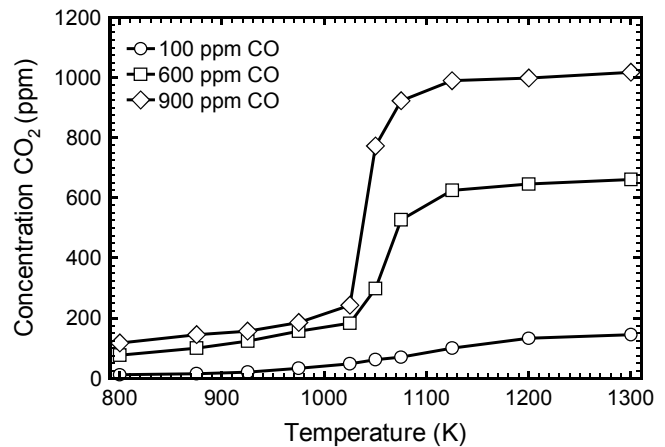


Figure 20. CO_2 concentration as a function of temperature for three concentrations of carbon monoxide for initial conditions of 420 ppm NO_2 , 760 ppm NH_3 , and 13.0% O_2 .

nitrogen appears to be impeded by the higher oxygen concentrations.

Figures 19 and 20 show the CO and CO_2 concentrations, respectively, as a function of temperature. For temperatures below about 1050 K, CO was found at concentrations slightly lower than the inlet concentrations with decreasing tendencies. For the same temperature range, CO_2 concentration was observed at values below 300 ppm. The CO concentration showed decreasing tendency between 800 and 1050 K and the CO_2 values showed the expected increase. Above 1050 K, CO concentrations decrease, while CO_2 concentrations increase.

The results from figures 19 and 20 (for the higher oxygen concentration) may be compared to figures 12 and 13 (for the lower oxygen concentration). This comparison indicates that the CO oxidation to CO_2 is enhanced at the higher oxygen concentration.

DISCUSSION

Ammonia Effect: To determine the effect of ammonia on nitrogen dioxide, results for the low oxygen concentration in figures 2 and 7 may be compared. As shown, ammonia has a significant effect on the conversion of NO_2 . Significantly lower concentrations of NO_2 were observed for the cases with NH_3 for the same temperature. For the case with ammonia (for low oxygen), essentially all NO_2 was removed for temperatures greater than about 1100 K; whereas for the case without ammonia, the temperature was greater than about 1200 K.

Ammonia also caused the production of N_2O , which could not be observed in significant

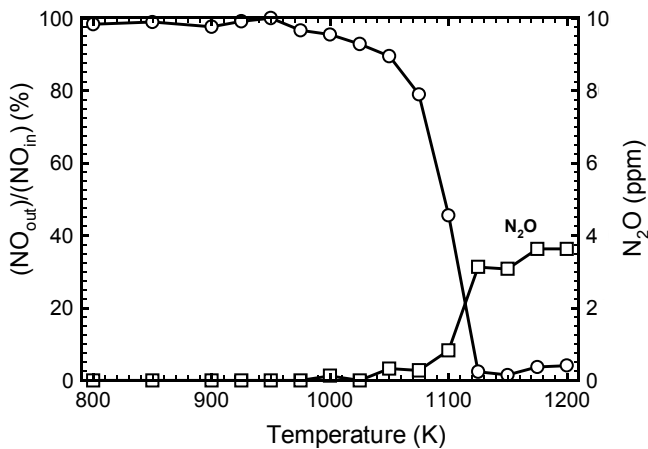


Figure 21. As adapted from [8], nitric oxide (NO) removal and N₂O emissions as functions of temperature for the SNCR process using ammonia for inlet concentrations of 330 ppm NO, 495 ppm NH₃, 1% oxygen, 0 ppm CO and 0% H₂O.

amounts for the “no ammonia” cases. Finally, the conversion of CO to CO₂ was strongly enhanced for the cases of ammonia present in the gas composition.

Another perspective of this work is to compare these results (for the conversion of NO₂) with results of for the more conventional SNCR process using ammonia which seeks to remove NO. By using results from previous work [8] for similar cases, a comparison can be completed. Figure 21 shows the normalized NO and the absolute N₂O concentrations as functions of temperature for the following conditions: 330 ppm of inlet NO, 495 ppm of ammonia and 1% oxygen with no CO or H₂O [8]. This case is similar to the case of figure 8 for NO₂ conversion.

This comparison indicates that the NO removal initiation temperature (about 1100 K) is similar in both cases. For higher temperatures for the NO₂ study, the removal of NO is only about 50%. Whereas for the more conventional SNCR process, the removal of NO is almost complete for the higher temperatures.

In addition, for the major by-product N₂O, the two cases are not similar. That is, for the case of the NO₂ experiment, N₂O is between 0 and about 25 ppm (figure 9); whereas, for the more normal SNCR process with NO, N₂O production is much more modest (on the order of less than 5 ppm). These results may suggest significant differences in the detail chemistry.

Oxygen Effect: A comparison of the results from the experiments for the low and high oxygen concentrations provided several observations. This comparison indicated that increasing oxygen

concentrations suppresses the conversion of NO₂ especially at lower temperatures. Complete conversion of NO₂ at increased temperature could not be found for the high oxygen case. For the low oxygen case, NO concentration showed similar results as the NO removal using ammonia with strongly increased conversion above 1075 K.

Nitrous oxide was observed to have two major dependencies on oxygen: (1) the overall concentration of N₂O increased with increasing oxygen concentrations, and (2) the 13.0% O₂ case (figure 16) showed a maximum N₂O concentration at about 1050 K, whereas for the low oxygen case (figure 9), the N₂O concentration did not exhibit such a maximum condition.

A strong effect of oxygen on the ammonia slip was also observed. At 800 K, the removal of NH₃ is visible for both studies with lower concentrations of NH₃ for 13.0% O₂ for the complete temperature range. The lowest temperature for complete NH₃ removal decreased from 1200 K (all cases for 0.73% O₂) to 1050 K (all cases for 13.0% O₂).

The conversion of CO to CO₂ could be observed for both studies, which lead to the conclusion that a significant effect on the CO concentration could be achieved with this process. As would be expected, higher oxygen concentrations were found to enhance this process to achieve significant conversion at lower temperatures.

Carbon Monoxide Effect: In the present study, it was found that carbon monoxide concentration had an effect on the chemistry leading to NO₂ conversion using ammonia. For the lower oxygen concentration, CO had a suppressive effect in terms of NO₂ conversion, whereas for high oxygen concentrations, CO enhanced the NO₂ conversion. The effect on NO production was inconclusive for low oxygen concentrations. For the case of 13.0% oxygen, lower CO concentration enhanced the production of NO at temperatures above 1050 K.

No effect of CO on the N₂O concentration was found for the low O₂ case. For high O₂ concentration, CO had the tendency to produce less N₂O. The effect of CO concentration on the ammonia exit concentration was a function of temperature. For lower temperature (in the case of 13.0% O₂, below 1050 K), CO suppressed NH₃ removal. For higher temperatures, CO was found to enhance the NH₃ removal by lowering the minimum temperature of significant NH₃ removal for low oxygen concentrations. A slight effect was

found in terms of CO conversion to CO₂, where low CO concentrations resulted in higher conversion temperatures for 0.73% O₂.

SUMMARY AND CONCLUSIONS

This investigation considered the removal of NO₂ in a SNCR process using ammonia. The main parameters which were varied were the temperature, and the carbon monoxide and oxygen concentrations.

- ◆ Ammonia promoted the conversion of NO₂ to NO as well as some direct reduction of NO₂ to N₂. For the cases with ammonia, greater levels of NO₂ conversion were observed for the low oxygen (0.73%) cases as compared to the high oxygen (13%) cases.
- ◆ For the cases with ammonia, the effect of CO concentrations was modest on NO₂ conversion.
- ◆ For the cases of no ammonia, increasing concentrations of CO tended to promote the conversion of NO₂.
- ◆ CO oxidation to CO₂ was enhanced in the presence of ammonia, and was further enhanced for the higher oxygen cases.
- ◆ No significant N₂O was detected for the cases without ammonia; but for the cases with ammonia, N₂O was detected up to levels of 50 ppm. The highest N₂O concentrations were observed for the higher oxygen cases with ammonia.
- ◆ Many of the global characteristics of this process were similar to the more conventional SNCR process based on removing NO. In particular, major changes were observed in species concentrations for temperatures beginning at about 1050 to 1100 K. One exception to the similarities was the production of N₂O which was greater for the NO₂ cases.

ACKNOWLEDGEMENTS

This work is supported in part by a grant from the Texas Advanced Research Program of the State of Texas under Grant No. 000512-0191-1999. The contents of this paper, however, do not necessarily reflect the opinions or views of the sponsors.

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Appendix A

The following table is a list of the specific wave numbers used to quantify each species, and the estimated lower detection limit. Lower values could be obtained, but the accuracy would not be as high.

For some of the species, more than one wave number was used. This helped avoid using a wave number which was obscured by other species (particularly H₂O and CO₂). Complete details of the calibration procedures and results are provided by Gentemann [7].

Species	Wave Number (cm ⁻¹)	Lower Limit (ppm)
NO	1900.6	5
CO	2166.0 2112.0 2162.4	5
CO ₂	2359.2 2336.4	5
N ₂ O	2213.8	2
NO ₂	1602.7	2
NH ₃	966.3 931.4 1084.8 868.0	5