NO_x Removal with Combined Selective Catalytic Reduction and Selective Noncatalytic Reduction: Pilot-Scale Test Results

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Pilot-scale tests were conducted to develop a combined nitrogen oxide (NO_x) reduction technology using both selective catalytic reduction (SCR) and selective noncatalytic reduction (SNCR). A commercially available vanadium— and titanium-based composite honeycomb catalyst and enhanced urea (NH₂CONH₂) were used with a natural-gas-fired furnace at a NO_x concentration of 110 ppm. Changes in SNCR chemical injection temperature and stoichiometry led to varying levels of post-furnace ammonia (NH₃), which acts as the reductant feed to the downstream SCR catalyst. The urea-based chemical could routinely achieve SNCR plus SCR total NO_x reductions of 85 percent with less than 3 ppm NH₃ slip at reductant/NO_x stoichiometries ranging from about 1.5 to 2.5 and SCR space velocities of 18,000 to 32,000 h-1. This pilot-scale research has shown that SNCR and SCR can be integrated to achieve high NO_x removal. SNCR provides high temperature reduction of NO_x followed by further removal of NO_x and minimization of NH₃ slip by a significantly downsized (high-space velocity) SCR.

Introduction

The 1990 Clean Air Act Amendment requirements for control of nitrogen oxides (NO_x) may be met by various control technologies depending on the combustion system application and the level of control needed. These technologies include selective catalytic reduction (SCR), which involves a heterogeneous reaction of NO_x and ammonia (NH_3) on the catalyst surface to form nitrogen (N_2) and water (H_2O) at temperatures typically ranging from 300 to 450 °C. These catalyst beds are often honeycomb or plate structures formed from metal oxides, zeolites, or noble metals. Oxides of the transition metals, including Co, Fe, Ni, V, and W, are usually most reactive at temperatures above 250 °C.

Implications

Continued research, development, and demonstration of the combined SCR/SNCR process will present an alternative technology for use in achieving NO_x reductions. This will expand the available control options and allow for staged implementation of technologies to achieve the mandates of the 1990 Clean Air Act Amendments. The successfully combined technologies emphasize the importance of structuring the regulations to allow maximum flexibility in the choice of compliance methods without restricting options to specific technologies. This further allows for advances and new developments in technologies to improve the performance of existing systems.

Both pilot- and field-scale results are available for SCR technology, showing NO_x removals above 90 percent. Most commercial SCR systems are designed to achieve 75 to 90 percent NO_x conversion.

SCR systems were installed at 250 commercial plants as of 1990, including 40 coal-fired applications totaling 11,000 MW(e) of capacity.3 In California, more than 75 gas turbine units are equipped with SCR systems. Key performance criteria affecting the design of a commercial SCR system are NO_x conversion, NH₃ emissions (NH_{3SLIP}), pressure drop, catalyst/system life, sulfur dioxide (SO₂) oxidation to sulfur trioxide (SO₃), and operating environments. Because these constraints can vary greatly from one installation to another, SCR catalysts of different physical and chemical properties have been developed to meet these commercial needs. The main concerns with respect to SCR technology include the potential for particulate fouling and catalyst poisoning by trace metals, SO2 and SO3. Particulate fouling and catalyst poisoning may reduce catalyst life, which is one of the most important factors in SCR economics. Catalyst poisoning occurs by reaction of solid, liquid, or gaseous matter with the catalyst or through formation of a masking layer over the surface of the catalyst that blocks diffusion of NO, and NH, to the available reactive sites.

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Selective noncatalytic reduction (SNCR), which involves hightemperature-furnace injection of a solid, gaseous, or liquid N-based reducing agent such as urea (NH2CONH2) or NH3, reacts homogeneously to convert NOx to N2. SNCR is a proven technology that has application to utility and industrial boilers burning a variety of fuels and waste-to-energy facilities. With this type of reduction, reaction temperatures range from 800 to 1100 °C. SNCR has achieved about 60 percent NO, reduction on a coalfired 150 MW(e) boiler2 at a molar ratio of reductant N to initial NO, concentration (normalized stoichiometric ratio or NSR) of 2,50 to 70 percent NO, reduction on a 135 MW(e) coal-fired boiler at an NSR of 1.2 to 1.63, and has been the subject of numerous laboratory or pilot-scale studies⁴⁻⁷. Pilot-scale laboratory studies have shown NO_x reduction around 60 to 80 percent depending on initial NO_x and chemical stoichiometry with levels of NH_{3SLIP} and nitrous oxide (N2O) below 5 and 20 ppm, respectively8.

The potential problems of using SNCR center around by-product emissions of NH₃ (NH_{3SLIP}), which result from incomplete reaction of the injected reductant. NH_{3SLIP} is generally caused by operation outside of the effective temperature range. NH_{3SLIP} can react with SO₃ to form ammonium bisulfate (NH₄HSO₄) around 275 °C and ammonium sulfate [(NH₄)₂SO₄] around 150 °C. The ammonium salts can form deposits on heattransfer surfaces causing reduced heat transfer and increased pressure drop. NH_{3SLIP} can also react with hydrogen chloride (HCl) or chlorine (Cl₂) to form ammonium chloride (NH₄Cl) around 100 °C resulting in a visible white plume in the stack emissions.

This research investigated the combination of SNCR and SCR technologies with the aim of demonstrating a more economical method of high NO_x removal. This "hybrid" process^{10, 11} aims to achieve combined NO, removal from the SNCR process initially and then further NO, removal from the SCR downstream. Because significant NO, reductions can be achieved by the upstream SNCR, downsizing of the SCR for only supplemental NO_x reduction becomes possible. The SCR catalyst utilizes the normally undesired NH3SLIP as the SCR reactant feed to further reduce NO_x as it minimizes NH_{3SLIP} problems. Additional NH₃ can be added upstream of the catalyst, if necessary. The smaller catalyst bed (higher space velocity) and elimination of reheat requirements will result in considerable cost savings through minimizing catalyst costs and equipment modification costs such as, reactor size, ductwork expansion, and fan size. The smaller catalyst volume may also reduce the potential for oxidation of SO₂ to SO₃ and subsequent formation of catalyst-plugging ammonium salts.

An adaptation of this technology has undergone full-scale testing by others¹² on a 110 MW gas/oil-fired boiler using in-duct and air preheater catalysts. SNCR-urea injection and SCR-ammonia injection resulted in NO_x reductions varying from 72 to 91 percent, depending on boiler load.

Pilot-scale work on a natural gas combustor examined the optimum combination of SNCR and SCR by varying SNCR injection temperature and chemical injection rate along with SCR space velocity and NH₃ feed rate. Emissions were monitored for NO_x, NH₃, N₂O, and combustion gases. The limited scope of work reported here discusses only one SNCR reductant chemical and SCR catalyst, although other chemicals and catalysts also were tested.

Experimental

SNCR/SCR tests were run on the U.S. Environmental Protection Agency's Innovative Furnace Reactor (IFR), a pilot-scale 14.7 kW (50,000 Btu/h), refractory lined, natural-gas-fired cylindrical furnace (see Figure 1). The IFR has an inner diameter of 15.2 cm and an overall vertical length of about 4 m. Several ports,

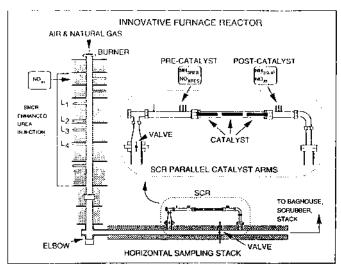


Figure 1. Schematic of the innovative furnace reactor (IFR) with the hybrid system.

spaced approximately 30 cm apart, are located along the vertical length of the furnace and can be used for sampling probes or chemical injection probes. The furnace simulates the gaseous combustion environment and quench-rate conditions anticipated in utility and industrial boilers. Typical operating concentrations were 110 ppm NO_x; dry measured values are adjusted for actual in-furnace water concentrations. The IFR total gas flow is 391 L/min. Reported values are at standard temperature, 0 °C, and pressure, 1 atm (STP), including an excess air of 50 percent. All monitored gases are adjusted to account for air in-leakage, most of which occurs near the elbow, resulting in typical operating oxygen (O₂) concentrations ranging from 5.1 to 6.5 percent and a NO_x concentration of about 95 ppm at the SCR.

Two 4.72 cm inner diameter stainless steel arms were added in parallel to the IFR horizontal sampling stack to hold the SCR catalysts. The tested catalyst was loaded into the catalyst arms in three 20.32 cm sections with about 2.5 cm spacing between each pair of sections. The flow from the furnace could be run through one or both of the catalyst arms, yielding two space velocities. Both catalyst arms were heat-traced and insulated to maintain a constant, monitored temperature of 315 °C.

Gas sampled from the horizontal sampling stack of the IFR was passed through sample lines to continuous-emission monitors (CEMs). Dry NO_x measurements were determined by a chemiluminescent method and reported as wet values at actual O₂ concentration. The reported NO_x concentrations do not include NO₂, because earlier tests showed that the NO₂ concentrations were below 5 percent of the total NO_x concentration.

Gas samples for carbon dioxide (CO₂), carbon monoxide (CO), and O₂ were first passed through a Hankison gas dryer and a desiccant canister of anhydrous calcium sulfate (CaSO₄). The desiccant was not used with the NO_x analyzer because CaSO₄ absorbs NO_x. All of the above on-line CEMs were zeroed and spanned with gases of known concentration both before and after each daily trial. N₂O concentrations were monitored by on-line gas chromatography (GC). The GC was used for analysis of grab samples taken before and during testing.

The concentrations of stack gas NH₃ were determined by two wet methods. The first was a batch method using a Fisher Accumet ion-selective electrode with a temperature-compensating probe. The second was a continuous-sampling system by Severins Science Instruments (SSI) that operates under the same principle. Both methods remove water for dry and semidry measurements, respectively. The first uses dual impingers in ice, and the second uses a room temperature condensing impinger. A comparison of

results between both methods showed excellent agreement when the SSI system was calibrated within a limited range of interest. The stack gas was drawn through an impinger system containing 0.02N H₂SO₄, and the pH was then adjusted with 10M NaOH solution. The NH₄ ion selective electrode, coupled with a pH meter, determines the NH₃ concentration. The meter and electrode were calibrated prior to analyses with known standards and checked throughout the testing to ensure that the values fell within the manufacturer's limits. NH₃ values, from both methods, were reported at actual H₂O and O₂ concentrations.

Urea Solution Injection

Testing during this work used injection of a proprietary ureabased solution with the trade name NO_xOUT A PLUS (Nalco Fuel Tech). NO_xOUT A PLUS, hereafter referred to as "enhanced urea," is a urea-based solution with antiscalants, dispersants, and more reactive ammonium compounds¹³ that shift the equilibrium point of the N₂O formation mechanism¹⁴ away from the N₂O product.

Different ratios of urea/NO_x were provided to the furnace by varying the concentration of the enhanced urea solution while maintaining a constant volumetric injection rate. A peristaltic pump was used to deliver low concentrations of reagent to the furnace. All tests proceeded with determination of stable baseline emissions during 100 percent H₂O injection to simulate the SNCR's supply of chemical. When baseline emissions stabilized, the chemical solution was injected at the desired urea/NO_{xi} stoichiometry. After completing emissions analyses, H₂O only was injected until the IFR returned to baseline emission concentrations.

Solutions were injected through a water-cooled probe that injected coaxially and coflowing to the process gas. The injection probe uses air (15 percent of the total IFR air flow) to effect droplet atomization. The droplet-size distribution exiting the nozzle was determined through the use of a Munhall particle size analyzer, which determines droplet size by measuring diffraction of laser light. These droplet-sizes were measured outside of the IFR using $\rm H_2O$ flow rates and pressures identical to in-furnace operation. The nozzle had a droplet-size distribution with a $\rm D_{50}$ of 13 μm and a $\rm D_{50}$ of 88 μm .

A commercial V₂O₅-based SCR catalyst, NO₂Cat™ VNX™, was used to test the catalytic reduction efficiency of NO, at 300 to 400 °C. This catalyst, supplied by Engelhard Corporation, is a composite formulation of V₂O₅/TiO₂ materials bonded to a ceramic honeycomb substrate. The catalyst specific area can be varied by using honeycombs of different cell density, ranging from = 4 to 31 cells per cm² (25 to 200 cells per inch²), for most industrial applications. Increasing cell density, which increases catalyst specific area, improves the volumetric rate for NO, reduction since the reaction is diffusion limited. However, it also increases the pressure drop at the same space velocity. Thus, a compact SCR system may be designed using catalyst honeycombs with high cell density. For this study, a 9.9 cells per cm² (64 cells per inch²) VNX™ catalyst was chosen for measuring SCR catalyst effectiveness at removing NO_x with enhanced-urea solution. Measured pressure drops were 1.8, 2.2, and 2.9 cm of water for 18,000, 24,000, and 32,000 hr-1 space velocity, respectively. The reductant was supplied solely by residual, unreacted NH₃ from the SNCR process. This NH₃ was measured prior to the catalyst inlet (see Figure 1) and termed NH3RES.

Temperature Profiles

The temperature profiles through the IFR were determined using a suction pyrometer and a type R thermocouple. Temperatures were determined during injection of air and H₂O to mimic the conditions expected during injection of the SNCR and SCR

reagents. The temperature at the point of the injection nozzle was calculated by linear extrapolation of the temperature values from downstream ports. The quench rate for natural-gas-firing with injection was nominally 114 and 240 °C/s at the low- and high-injection ports, respectively.

Tests were conducted at a limited range of operating conditions by varying the SNCR injection port level (T_{INI}), the SCR gas-flow rate or space velocity (SV), and the amount of chemical injection. Operating conditions were chosen based on earlier work¹⁵ that had defined the general system performance over a broad range of T_{INI}, SV, and NSR. The enhanced urea feed rate into the SNCR system is called the Normalized Stoichiometric Ratio (NSR) and is a ratio of the available N reductant groups to the baseline NO_x concentration NO_{xi}. Urea-based chemicals decompose into 2 moles of reductant N per mole of urea. The value of NSR is calculated by multiplying the concentration of enhanced urea solution (moles N/mL) by the solution feed rate (10 mL/min) then dividing by the measured NO_{xi}. The NH₃ feed into the SCR system is supplied solely by the unreacted NH₃ from the SNCR process, NH_{3RES}. The rate of feed is termed SR_{RES} where

$$SR_{RES} = \frac{NH_{3RES}}{NO_{3RES}} . {1}$$

Here, NH_{3RES} refers to the amount of residual NH_3 reductant prior to the catalyst, and NO_{xRES} is the amount of NO_x residual after SNCR and prior to the catalyst. SR_{RES} is determined from the sampled NH_3 concentration prior to the catalyst.

SCR tests were first conducted with enhanced urea injection to establish the baseline performance characteristics. These tests were rerun at the end of the test matrix and ensured that no degradation in catalyst performance had occurred.

Results and Discussion

Figure 2 shows the general optimum operating range of an SNCR system for NO_x removal (X_{SNCR}) as a function of T_{INJ} and NSR. The contour plot fit to the data shows that between 850 and 950 °C, NO_x removals of 60 to 75 percent were achieved at an NSR of greater than 1.5 ($NO_{xi} = 110$ ppm). A few chemical injections were made beyond the optimum SNCR injection-temperature window, simulating sudden load swings in a full-size plant. At higher T_{INJ} cases, the enhanced-urea chemical will undergo partial oxidation, creating NO_x . This would create more

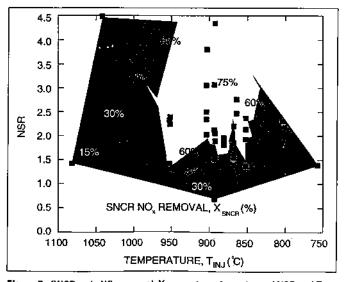


Figure 2. SNCR-only NO_x removal, X_{SNCR}, at varying values of NSR and T_{INJ}. Shaded regions model the data points (filled squares) over the experimental domain.

of a NO_x burden on the downstream SCR. At injection temperatures below the optimum, considerable reaction inefficiency is expected, leading to lower values of NO_x reduction and high levels of NH_{3RES} . In either the high- or low-injection temperature case, the X_{SNCR} values shown on Figure 2 are considerably lower than at the middle injection temperatures.

Figure 3 illustrates the NSR and $T_{\rm INI}$ conditions under which NH_{3RES} from the SNCR operation provides the reductant for the downstream SCR operation. NH_{3RES} is expressed here as a ratio with NO_{xRES} (the residual NO_x prior to the SCR that was unreduced during SNCR) or the SCR stoichiometry, SR_{RES}. Figures 2 and 3 are printed with the same ranges to facilitate direct comparison. Because NH₃ measurements were not made for all of the tests, Figure 3 will have less data points than Figure 2. The majority of the tested SNCR conditions provided a functional reductant to NO_x ratio, approximately 0.4 to 1.2, into the SCR system without the necessity of additional precatalyst reductant feed.

The NO_x removal performance of the SCR system (X_{SCR}) is shown in Figure 4 plotted against SR_{RES}. The reductant chemical for the SCR, NH_{3RES}, is supplied solely from the enhanced urea

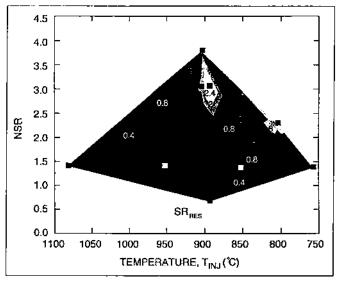


Figure 3. Pre-SCR stoichiometry, the ratio of residual NH_3 to residual NO_x ($NH_{3RES}/NO_{xRES}=SR_{RES}$), resulting from SNCR operation at varying values of NSR and T_{INJ} . Shaded regions model the data points (filled and open squares) over the experimental domain.

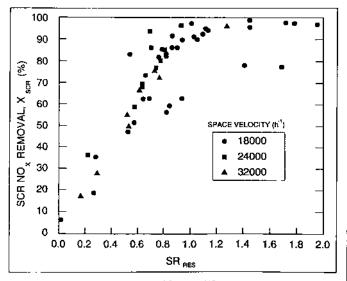


Figure 4. Effect of SR_{RES} and SV on SCR-onty NO $_{x}$ removal (X_{SCR}), (NO $_{xRES}$ = 23 to 662 ppm).

injected with the SNCR operation. Prior SCR-only testing with enhanced urea fed into the endplate, without SNCR injection, showed no distinguishable performance differences from when the reductant was supplied solely by NH_{3RES} from SNCR operation. This implies that the residual reductant species from the SNCR enhanced urea chemical performs similarly to NH₃ injected ahead of the catalyst bed, regardless of the SNCR T_{INI}.

Figure 4 indicates that SCR NO_x removal is a function of stoichiometry, SR_{RES}; values of SR_{RES} approaching 1.0 led to NO_x removals above 90 percent, in most cases. While space velocity was varied from 18,000 to 32,000 hr-1 during the testing, no obvious effect was observed. This likely results because under these high-conversion, low-NH_{3SLIP} conditions, the catalyst NO_x conversion is limited by available NH₃, (or SR_{RES}) not by residence time in the SCR. However, as the SV increases further, the NO_x conversion will begin to decrease with increasing flow rate as the NH₃ has insufficient time to diffuse and react.

The SCR NO_x removal can be compared with its NH_{3SLIP} results, Figure 5. In the SR_{RES} range from 0.9 to 1.0, Figure 5 shows that NH_{3LSIP} starts to increase above 5 ppm. Figure 4 shows that X_{SCR} has levelled off at above 90 percent removal. The data show that the NH_{3SLIP} performance of the SCR, at least below a stoichiometry of unity, is apparently independent of both space velocity (SV) and the input NO_x value, NO_{xRES} .

Hybrid SCR/SNCR

The NO_x and NH_{3SLIP} performance results of the combined SNCR and SCR systems are shown in Figure 6. Because of the large amount of NH_{3SLIP} values less than 1 ppm, the ordinate is plotted on a logarithmic scale for clarity. All of the data at ≤ 85 percent total NO_x reduction had less than 1 ppm NH_{3SLIP} . Because the data represent varying test conditions of NSR and T_{INJ} , both optimal and suboptimal operating conditions are shown. This illustrates the importance of proper operation, monitoring, and control of the SNCR system toward achieving consistently optimum hybrid process NO_x and NH_{3SLIP} performance. Although Figure 6 also indicates variation of SV (18,000 to 32,000 hr-1), no relation to NO_x reduction or NH_{3SLIP} results was apparent with the limited number of tests conducted.

Figure 7 plots the percentage of unconverted NH₃ against SR_{RES} where the former is defined by

$$NH_{3UNC}(\%) = \frac{NH_{3SLIP}}{NH_{3RES}} *100.$$
 (2)

This is similar to Figure 5, but now also discriminates the data by hybrid versus SCR-only operation. All of the results show less

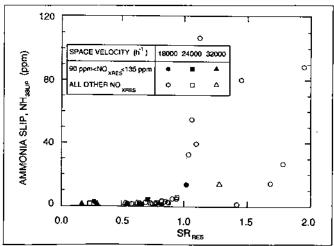


Figure 5. Effect of SR_{RES}, SV, and NO_{xRES} on NH_{3SLIP} in the hybrid system. (NO_{xRES} = 25 to 463 ppm).

than 5 percent unconverted ammonia until SR_{RES} values, exceeding about 0.9, are reached. No distinguishable dependency on NO_{xRES} can be noted. No apparent difference in catalyst function is noted between the SCR-only results and the hybrid results.

While not the main focus of this work, measurements of preand post-SCR N₂O showed concentrations that were always below 7 ppm. Baseline CO was always below 15 ppm, increasing to 27 ppm during testing.

Optimization of the hybrid SCR/SNCR system performance must take into account a balance between NO_x reduction and NH_{3SLIP}. The latter can be illustrated by combining Equations 1 and 2 into a definition of NH_{3SLIP}:

$$NH_{3SLIP} = NH_{3UNC} \cdot SR_{RES} \cdot NO_{XRES}$$
 (3)

where

$$NH_{3UNC} = f$$
 (cat.type, SV, cat. T). (4)

Equation 3 shows that NH_{3SLIP} can be minimized by reducing either the SR_{RES} or NO_{xRES} term. Thus, if either lower NO_{xRES} reaches the SCR or the SCR can be operated at a lower SR_{RES} value — still achieving the target overall NO_x reduction — then the amount of NH_{3SLIP} will be reduced.

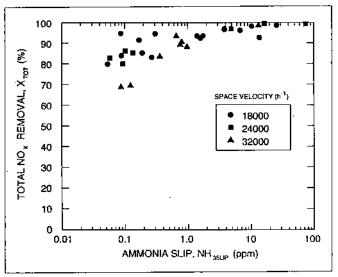


Figure 6. Combined-hybrid system performance: total NO $_{\rm x}$ removat and NH $_{\rm 3}$ slip. (756 °C < T $_{\rm INJ}$ < 952 °C, NO $_{\rm xi}$ = 103 to 126 ppm).

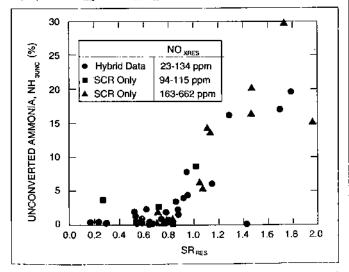


Figure 7. Effect of SR_{RES} on unconverted NH3, NH3UNC, for three ranges of NO $_{\text{XRES}}$

Minimization of both NO_{xRES} and SR_{RES} depends on high levels of SNCR reagent utilization achieving significant reductions in NO_{xi} (lowering NO_{xRES}) with sufficient, yet not excessive, amounts of unconverted reductant (NH_{3RES}) passed on to the catalyst. Optimization of the hybrid system is achieved when the SNCR technology is operated at peak performance, maximizing NO_x reduction while achieving a NH_{3RES} value that results in an SR_{RES} value into the catalyst that will not result in excessive NH_{3SLIP}.

This concept is developed as follows for a facility with a hypothetical limit of 3 ppm NH_{3SLIP} and a $NO_{xi} = 110$ ppm. First, for a range of assumed SNCR NOx reductions, the maximum allowable unconverted ammonia, NH3UNC, or 3 ppm * 100 / NO_{xRES}, is associated with its experimentally determined SR_{RES} from Figure 7. This value of SR_{RES} is the highest stoichiometry at which the SCR can operate without exceeding 3 ppm NH_{3SLIP}. The anticipated SCR NO_x reduction at this SR_{RES} value is determined from previous experimental results reported in Figure 4. Finally, these results are plotted in Figure 8 showing the optimal performance of the SCR/SNCR system given an assumed effectiveness of the SNCR process and a limit of 3 ppm NH_{3SLI}, shown using the solid line on Figure 8. Of course, these results are a strong function of the SCR's performance (see Equation 4) and the initial NO, level in the facility. The latter is an issue because, for a given SNCR percentage NO_x reduction, as NO_{xi} increases the fraction of allowable unconverted ammonia decreases. This results in lower tolerable levels of SR_{RES} and, therefore, lower SCR NO, reduction. Full-scale applications may achieve lower performance because of reactant-mixing limitations causing non-uniform reagent distributions. Use of computational fluid dynamics and kinetic modeling, coupled with appropriate injector and flow design criteria, can ameliorate these limitations.

Our results can be extrapolated to higher NO_{xi} systems, given the assumptions:

- SCR NO_x reduction performance, at a given value of SR_{RES}, is not sensitive to NO_{xi} and can therefore be approximated by a curve fit to Figure 4; and
- SCR NH_{3UNC} performance, at a given value of SR_{RES}, is not sensitive to NO_{xi} and can be approximated by a curve fit to Figure 7.

Both of these assumptions are validated by the results of this research over the range 23 ppm $\leq NO_{xi} \leq 662$ ppm. The SCR

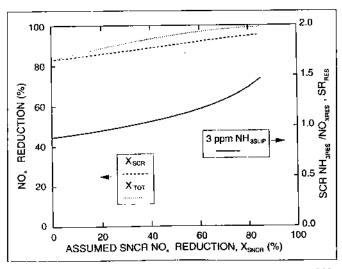


Figure 8. Optimal-operating conditions and performance for the hybrid SCR/SNCR system at NO $_{\rm xi}$ = 110 ppm based on the maximum allowable SR_{RES} that will not exceed 3 ppm NH $_{\rm 3SLP}$ From results presented in Figures 4 and 7.

system's NO_x reduction performance had no apparent dependency on NO_{xi}, which is not shown, and Figure 7 indicates that NH_{30NC} performance was insensitive to NO_{xi} over the same range.

For a system with NO_{xi} = 500 ppm, the results in Figure 9 show a higher incremental benefit of the hybrid system (versus SCR alone) than in Figure 8. NO_x reduction increases from about 50 percent with the SCR alone to over 90 percent when SNCR achieves 65 percent reduction.

Figure 10 shows the total NO_x reduction results for NO_{xi} ranging from 110 to 500 ppm. Higher NO_{xi} values result in lower X_{TOT} at a given value of X_{SNCR} . At $X_{SNCR} = 0$, the maximum NO_x reduction performance of the SCR system (3 ppm NH_{3SLIP}) is observed. This observed NO_x reduction can be improved by adding the SNCR process, especially at higher NO_{xi} values where the slopes of the curves are steeper.

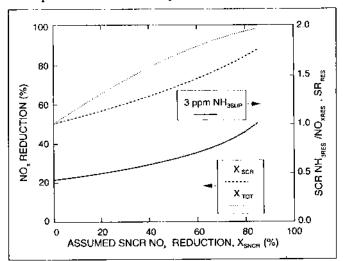


Figure 9. Optimal-operating conditions and performance for the hybrid SCR/ SNCR system at NO $_{\rm xi}$ = 500 ppm based on the maximum allowable SR_{RES} that will not exceed 3 ppm NH $_{\rm 3SLiP}$. From results presented in Figures 4 and 7.

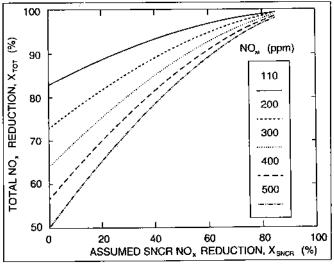


Figure 10. Total NO_x reduction for the hybrid system at several values of NO_{x1} based on results presented in Figures 4 and 7 and a limit of $NH_{3SLIP} = 3$ ppm.

Conclusions

Research on a pilot-scale combustor has shown that a hybrid SCR and SNCR process can achieve ≥ 85 percent NO_x reduction with ≤ 3 ppm NH_{3SLP} at a relatively high-space velocity (18,000 to 32,000 hr⁻¹). This hybrid system can be operated such that intentional NH_{3RES} from SNCR injection of an enhanced-urea chemical acts as a reductant feed to supplant or supplement the

SCR reductant. NO_x removals from the hybrid system are always higher than can be achieved by either SCR or SNCR alone at a given value of NH_{3SLIP}. The use of a high-space velocity SCR system indicates that a significantly downsized catalyst is sufficient for this hybrid system. Optimization of the hybrid system performance requires maximizing NO_x removal in the SNCR process. An analysis based on the hybrid system performance in this pilot-scale work indicates that a facility with NO_{xi} = 500 ppm will achieve a total NO_x removal of 90 percent with NH_{3SLIP} \leq 3 ppm only if the SNCR NO_x reduction is at least 65 percent.

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Nomenclature

D_x	Particle size below which x weight percentage of the sample resides, µm
NH _{3RES}	Sampled precatalyst NH ₃ concentration, ppm
NH _{3SLIP}	Sampled postcatalyst NH ₃ concentration, ppm
NOrt	Final NO, concentration, ppm
NO _x	Initial furnace NO, concentration, ppm
NO _{xRES}	Precatalyst NO, concentration, ppm
NSR	SNCR normalized-stoichiometric ratio, or the molar
	ratio of reductant N fed to NO _{xi} concentration, unitless
SRRES	Sampled-molar ratio of NH _{3RES} to NO _{3RES} , unitless
sv	Catalyst space velocity or gas-flow rate through
	catalyst divided by the catalyst volume at STP, hr-1.
X _{SCR}	Reduction of NO _x concentration in the SCR system,
	$[(NO_{xRES} - NO_{xi})/NO_{xRES}]*100$, percent
X _{SNCR}	Reduction of NO, concentration in the SNCR system,
	$[NO_{xi} - NO_{xRES})/NO_{xi}]*100$, percent
X _{TOT}	Total SCR and SNCR NO, reduction, [(NOxi - NOxf)/
	NO _{si}]*100, percent

References

- J. C. Smith, "Selective Catalytic Reduction (SCR) Controls to Abate NO_x Emission," Industrial Gas Cleaning Institute, Inc., Washington, DC, 1991.
- J. E. Hofmann, J. von Bergmann, D. Bökenbrink, K. Hein, "NO_x control in a brown coal-fired utility boiler, "Proceedings: 1989 Joint Symposium on Stationary Combustion NO_x Control, Vol. 2, U.S. Environmental Protection Agency, EPA-600/9-89-062b (NTIS PB89-220537), (1989).
- M. Jødal, T. V. Lauridsen, K. Dam-Johansen, "NO_x removal on a coalfired utility boiler by selective non-catalytic reduction," *Environ. Progress* 11(4): 296 (1992).
- L. J. Muzio, R. M. Himes, R. E. Thompson, "Simultaneous NO_x/SO Removal by the Dry Injection of Lime-Urea/Hydrate", U.S. Department of Energy, DOE/PC/88860-T1 (DE90009566), (1990).
- R. Mangal, M. Mozes, R. Gaikwad, R. Thampi, D. MacDonald, "Additive/injection for NOx control". Proceedings: 1989 International Symposium on Combustion in Industrial Furnaces and Boilers, American Flame Research Committee, Short Hills, NJ (1989).
- P. Teixeira, L. J. Muzio, T. A. Montgomery, G. C. Quartucy, T. D. Martz, "Widening the urea temperature window," Proceedings: 1991 Joint Symposium on Stationary Combustion NO_x Control, Washington, DC (1991) Vol. 3, U.S. Environmental Protection Agency EPA-600/R-92-093c (NTIS PB93-212868), (1992).
- R. Mangal, M. S. Mozes, P. L. Feldman, K. S. Kumar, "Ontario Hydro's SONOX process for controlling acid gas emissions," I Proceedings: 1991 Joint Symposium on Stationary Combustion NO₂Control, Washington, DC (March 25-28, 1991) Vol. 3, U.S. Environmental Protection Agency, EPA-600/R-92-093c (NTIS PB93-212868), (1992).
- B. K. Gullett, K. R. Bruce, W. F. Hansen, J. E. Hofmann, "Sorbent/urea slurry injection for simultaneous SO₂/NO_x removal," *Environ. Progress* 11(2): 155 (1992).

- B. E. Hurst, "Improved deNO_x process for coal-fired utility boilers," Presented at 11th Annual Stack Gas/Coal Utilization Meeting, Exxon Engineering Publication, Paducah, KY (1983).
- J. E. Hofmann, W. H. Sun, B. K. Luftglass, U.S. Patent No. 4,978,514, 1990.
- B. K. Luftglass, W. H. Sun, J. E. Hofmann, U.S. Patent No. 5,139,754, 1992.
- H. V. Krigmont, P. L. Chien, W. H. Pollock, J. J. Nylander, B. J. Williams, L. J. Czarnecki, L. B. Aller, "Full scale demonstration of Wahlco staged NO_x reduction system," Presented at 1993 Joint EPA/ EPRI Symposium on Stationary Combustion NOx Control, Bal Harbour, FL (1993).
- M. L. Lin, D. V. Diep, "NO_xOUT PLUS An Improved SNCR process for NO_x emission control," presented at AIChE Annual Meeting, Miami Beach, FL (1992).
- L. J. Muzio, T. A. Montgomery, G. C. Quartucy, J. A. Cole, J. C. Kramlich, "N₂O formation in selective non-catalytic NO_x reduction processes," Proceedings: 1991 Joint Symposium on Stationary Combustion NO_x Control, Washington, DC (1991) Vol. 2, U.S. Environmental Protection Agency, EPA-600/R-92-093b (NTIS PB93-212850), (1992).

 B. K. Gullett, P. W. Groff, M. L. Lin, J. M. Chen, "Pilot-scale testing of NO_x removal with combined selective catalytic reduction and selective non-catalytic reduction," presented at: 1993 Joint EPA/EPRI Symposium on Stationary Combustion NOx Control, Bal Harbour, FL (1993).

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