Add-on control techniques for nitrogen oxide emissions during municipal waste combustion

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Abstract

Throughout history, man has disposed of 'municipal solid waste' via incineration and landfilling. As the population and waste generated per capita increased, and concomitantly the available landfill space diminished, the favored and sometimes only waste disposal option was incineration.

Air pollution is one of the costs society bears for incineration. Municipal waste combustion liberates acid gas emissions such as SO_x, HCl, and NO_x. Flue gas scrubbing and alkaline sorbent injection technologies are typically used to neutralize acidic, SO_x and HCl emissions. Staged combustion and more frequently add-on, 'post-combustion' control technologies are used to reduce nitrogen oxide emissions.

This section of the journal will cover the use of 'add-on control technologies' for reducing nitrogen oxide emissions from municipal waste incinerators. A detailed technology description along with process benefits and limitations are discussed. Finally, a generic economic analysis and a list of post-combustion NO_x control applications are enclosed.

1. Introduction

Throughout the world, incineration is used to reduce the volume of waste required for disposal. Acidic SO_x, HCl, and NO_x bearing flue gases result from municipal waste combustion. Nitrogen oxide emissions are generated via high-temperature oxidation of fuel bound nitrogen and nitrogen contained in the combustion air. Since options for combustion NO_x controls are very limited for municipal waste incinerators and the reaction conditions for SNCR are ideal, post-combustion air pollution control technologies have emerged as the most cost-effective method for reducing NO_x emissions. This section of the journal will focus on the theory, performance,

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and economics of applying post-combustion NO_x control technologies to municipal waste incinerators.

2. Theory

Broadly, post-combustion NO_x control technologies can be classified as selective non-catalytic reduction and selective catalytic reduction technologies.

2.1. Selective non-catalytic reduction

Selective non-catalytic reduction (SNCR) can simply be defined as the injection of a selected nitrogenous compound into the hot flue gases of a stationary combustion source within the proper temperature range for reaction to yield nitrogen, water, and carbon dioxide. SNCR requires no catalyst and is selective in that reagents react with NO_x only, excluding all other flue gas components.

SNCR was developed and patented in the 1970s. Early applications were limited to anhydrous or aqueous ammonia injection (hazardous reagents) and suffered from high carrier gas requirements and a narrow reagent injection temperature range. Process improvements including the use of non-hazardous (stabilized) urea were developed and patented and are now marketed under the trade name NOxOUT®. The overall chemical reaction using urea is

$$NH_2CONH_2 + 2NO + 1/2O_2 \rightarrow 2N_2 + CO_2 + 2H_2O.$$
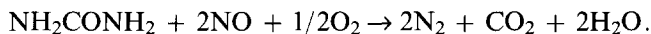
The principle components of an SNCR system are a reagent storage and injection system which includes tanks, pumps, injectors, and associated controls, and (optionally) a continuous emission monitoring system capable of monitoring NO_x and NH_3 (see Fig. 1). Given the simplicity of these components, installation of an SNCR system is relatively easy and retrofits do not require extended source shutdowns.

The NOxOUT SNCR process can reduce stack NO_x up to 90%, although typical reductions range from 50–75%. The process operates within a temperature range of 1400–2200 °F at >0.3 s residence time. In SNCR processes, there is a range of temperatures where significant NO_x reductions are obtained, defined as the 'temperature window' (see Fig. 2). Within this window, the NO_x reduction versus temperature curve consists of three zones: left side, right side, and plateau. This shape is the result of chemical kinetics. At temperatures lower than this window, the reaction requires longer reaction time than is typically available in most commercial combustion systems. Thus, NO_x reductions are negligible and by-product emissions (ammonia slip) are high. At the left side of the curve, NO_x reduction increases with increasing temperature; ammonia slip is still significant. At the plateau, reaction rates are optimum for NO_x reduction; ammonia slip is decreasing. A temperature variation in this zone has only a small effect on NO_x. A further increase in temperature beyond the plateau decreases NO_x reduction. At the right side, the oxidation of urea to NO_x becomes a significant reaction path and competes with NO_x reduction reactions for reagent. These oxidation reactions continue to increase with increasing temperature to the
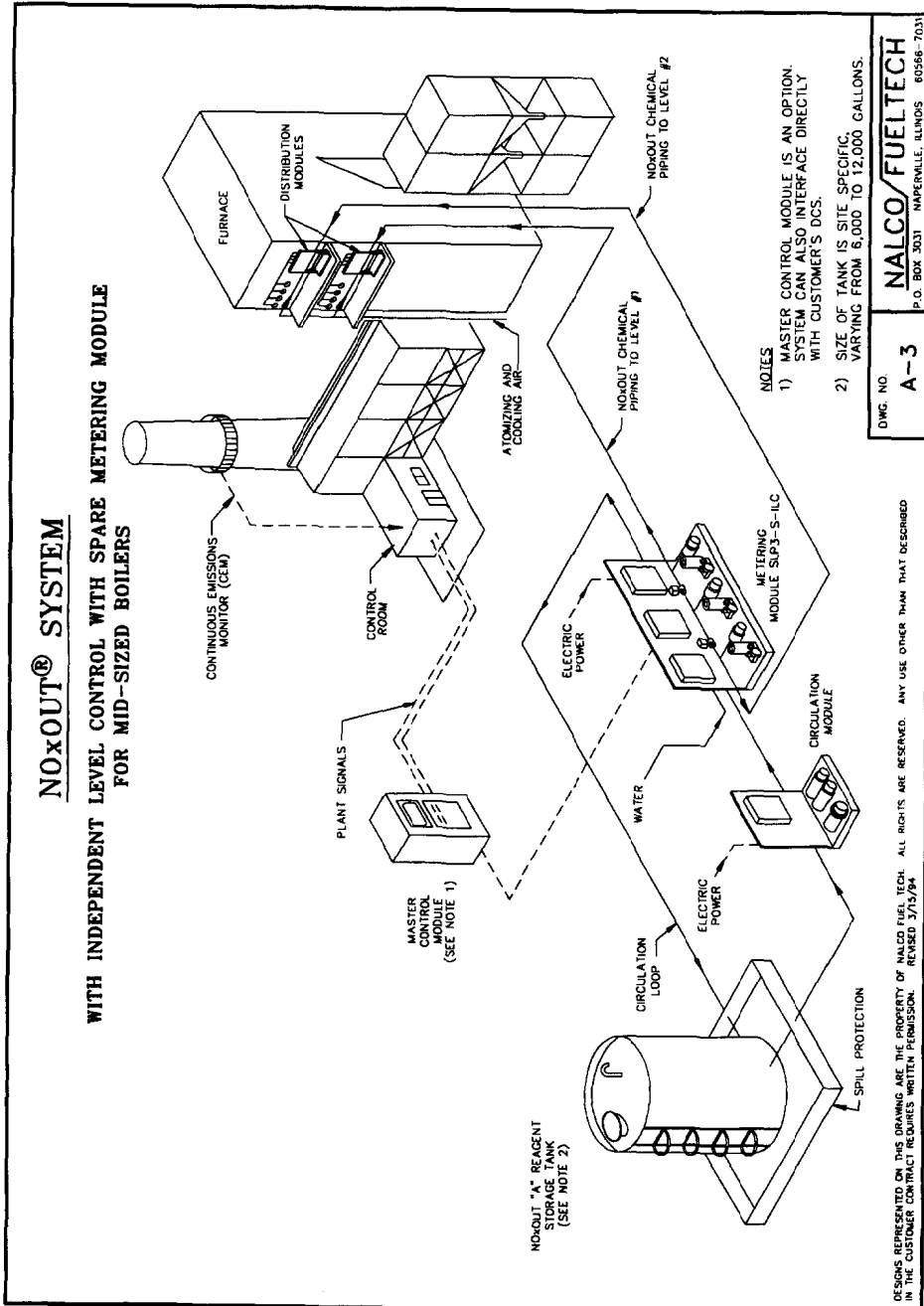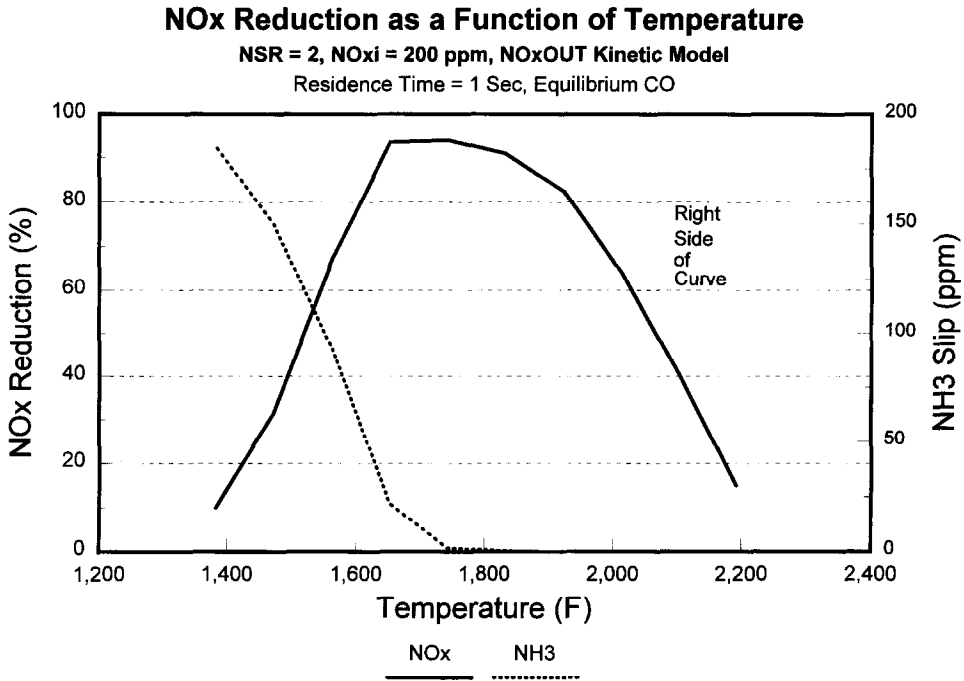
Fig. 1.

NOx Reduction as a Function of Temperature
NSR = 2, NOxi = 200 ppm, NOxOUT Kinetic Model
Residence Time = 1 Sec, Equilibrium CO

Fig. 2.

point of decreasing NO_x reduction while almost eliminating slip. A further increase in temperature beyond the right side increases NO_x above the baseline value. Although the reduction is less than the maximum, operation at the right side is practiced and recommended since by-product emissions decrease with increasing temperature.

The NO_x removal efficiency is related to a variable known as normalized stoichiometric ratio (NSR). The NSR is used as a measure of the rate at which urea is added to the flue gas relative to the baseline NO_x and is defined as

$$NSR = \frac{\text{actual mole ratio of urea to baseline } NO_x}{\text{theoretical mole ratio to reduce one mole of } NO_x}.$$

For urea, the theoretical mole ratio is 0.5 arising from the stoichiometry of the chemical reaction shown previously.

In addition to using a non-hazardous, non-toxic reagent, the NOxOUT Process (urea-based SNCR) has the following other advantages compared to first generation ammonia-based SNCR:

- Potentially lower capital cost due to the lack of large system compressors and elimination of anhydrous ammonia storage, handling, and safety equipment.
- Lower operating costs resulting primarily from minimization of distribution or atomizing gas (steam or compressed air) requirements.

- Inherently, more effective control of spray patterns and chemical distribution for better mixing with the use of liquid rather than gas-based reagents, thereby resulting in better chemical utilization.
- Chemical enhancers which can be used to improve control of potential by-product generation while reducing the NO_x concentration over an expanded temperature range.

Performance design tools increase confidence in applying NOxOUT to new applications. Process performance is analyzed using Nalco Fuel Tech's chemical kinetics computer model (CKM). Process conditions are evaluated using computational fluid dynamics (CFD) modelling techniques. The CFD modelling also enables the simulation of injector design configurations to evaluate chemical dispersion predictions of expected performance.[1]

2.2. Selective catalytic reduction

Selective catalytic reduction (SCR) is the other post-combustion, add-on technique for reducing municipal waste incinerator nitrogen oxide emissions. SCR employs the injection of ammonia as the flue gases passes through a catalyst bed of metal oxide, zeolite, or other suitable material. The application temperature window ranges from 550 to 1000 °F. SCR is capable of consistently achieving greater

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[1] Model descriptions: Flow modelling is performed using the PHOENICS CFD program (CHAM), with process-specific correlations provided through subroutines proprietary to Nalco Fuel Tech. The proprietary subroutines perform multi-component droplet dynamics calculations, provide physical property correlations and transport coefficient estimates, and perform supplemental computational or visualization functions.

Field Test data are used whenever possible to verify the model. Actual performance is often different than the design. Direct temperature measurements reduce uncertainties that can arise from assumptions or estimates of such factors as furnace wall cleanliness, gas emissivity, and fuel characteristics.

For a given set of operating conditions, the CFD model provides an estimate of the temperature of the flue gas as a function of residence time. This residence time versus temperature profile is the basis for predicting chemical process performance for a specific unit. Temperature estimates were not required for this project because the unit is nearly isothermal in the injection region.

The chemistry of the NOxOUT Process involves over 90 individual chemical reactions describing interactions among 31 species. To understand the effects of process parameters, a chemical kinetic model was developed. This model describes an ideal plug flow, i.e. no temperature or species concentration gradient in the radial direction and no back-mixing.

Reaction rates, density, and thermodynamic information are supplied by a library of gas-phase subroutines called CHEMKIN developed at the Sandia National Laboratories. This package of subroutines compiles user supplied reaction sets and a thermodynamic data file.

The CKM identifies the temperature ranges for effective NO_x control and provides estimates of the expected NO_x reduction and by-product emissions.

Injector simulation is performed by incorporating multicomponent droplet dynamics calculations into the CFD model. Sprays are modelled as sources of many individual droplets grouped into several classes. Each droplet class has a different droplet size, velocity, or angle relative to the principle spray direction. The mass, energy, and momentum sources of the injectors are included into the CFD model, and additional iterations were performed until the CFD and injector results converged to a steady-state solution.

than 80% NO_x reduction, the highest of the currently commercialized NO_x reduction methods. The primary disadvantage of SCR technology is cost. The high capital cost is associated with the catalyst and the construction of the reactor to house the catalyst bed. The relatively high operating costs are attributable to periodic catalyst replacement and disposal of spent catalyst as a hazardous waste. Also contributing to the operating costs are chemical reagents and system pressure drop. When the municipal waste incinerator utilizes a waste recovery boiler, an SCR system cannot be installed hot-side (immediately downstream of the economizer) as in a power boiler because of the high level of impurities in the gas that will poison the catalyst. However, the SCR system can be installed cold-side (after gas cleaning equipment) with reheating of the gas to achieve proper reaction temperature.

Fly ash deposition and other impurities can blind or poison up to one-third of the catalyst active area. Installation of the reactor after a wet flue gas desulfurization scrubber (cold-side) eliminates the catalyst blinding problems, but at the expense of up to a 3% heat rate penalty for reheating the flue gas to reaction temperatures. The in-line catalyst bed typically results in a 3″ water column pressure drop that must be overcome by the induced draft fan.

Despite the high capital and O & M costs, SCR is being installed on a small scale in Europe and Asia. To improve the economics of applying SCR, lower-temperature catalysts are being developed to eliminate reheat, as well as combination catalysts capable of oxidizing carbon monoxide and dioxins.

3. Discussion

The cost effectiveness of SNCR (particularly urea-based SNCR) has led many regulatory agencies to define it as the 'best demonstrated technology' for municipal waste incinerators. The data in Table 1 shows that significant reductions in NO_x can be achieved in municipal waste incinerators, ranging from 50% to 80%. In terms of concentration, this means that NO_x can typically be controlled to levels of less than 100 ppmvd (@7% O_2). The generally excellent performance that can be achieved in municipal waste incinerators is the result of several factors. Of most importance, the post-combustion gases will typically have residence times of one or more seconds in the range of 1500–1900 °F. These conditions are ideal for SNCR processes. Also, the concentration of carbon monoxide in the furnace tends to be slightly higher than thermodynamic equilibrium values which helps the reaction kinetics and will tend to minimize NH_3 slip.

Differences in NO_x reduction performance between individual applications can be ascribed to several factors:
(1) Fuel type,
(2) Initial NO_x concentration,
(3) Flue gas excess O_2,
(4) Flue gas CO concentration,
(5) Furnace geometry,
(6) Gas velocities and distribution,

Table 1
Municipal waste combustors

Company/Location	Unit type	Size (MMBtu/h)	Fuel	NO_x baseline (ppm)	Reduction (%)	Ammonia slip (ppm)
New Hanover County Wrightsville Beach, NC	Volund MWC	108	MSW	300	60	<15
Hamm (4 units) Germany	Moving Grate	528	MSW	170	41	<5
Herten (2 units) Germany	Moving Grate	242	MSW	185	60	<7
Frankfurt (4 units) Germany	Moving Grate	660	MSW	170	70	17
SEMASS Rochester, MA	Riley Stoker	375	MSW	220	50	<20
Falls Township Falls Township, PA	B & W Stoker	(2) 325	MSW	330 Max 285 Typ	50% Max 40% Typ	<11
Emmenspitz Zuchwil, Switzerland (D)	Moving Grate Incinerator	121	MSW	200	68	<20
(D)	Detroit Stoker	137.5	MSW	110	60	<10
Wheelabrator Millbury, MA (D)	Moving Grate Incinerator	325	MSW	240 dry 7% O_2	65	<15
City of Berlin Berlin, Germany (D)	Moving Grate		MSW	160	69	<25
(D)	Zurn Stoker	167	MSW	275	75	<6
Tekniskaverken Garstad (D)	Moving Grate		MSW			
Lisbon, CT				285 Typ	40% Typ	

(D) denotes demonstration not a commercial application.

(7) Furnace temperature profile,
(8) Load changes.
The most important of these are residence time and temperature. Newer units tend to have residence times greater than 1 s in the preferred temperature range. The initial NO_x concentration has an effect on NO_x reduction that can be expressed on a percentage basis, i.e. NO_x reduction capability typically increases with increasing NO_x baseline. Finally, an additional factor is that during most SNCR demonstrations, chemical injection was achieved through existing furnace penetrations. These penetrations may not have been optimally located to achieve maximum performance.

As stated earlier, Nalco Fuel Tech uses CFD and CKM computer modelling to select the optimum reagent injection strategy and boiler penetration locations. These models are also used to predict NO_x reduction performance and potential NH_3 slip.

The ultimate fate of NH_3 slip has not been fully described. Most of the measurements reported herein were based on extracting samples of the flue gas stream at a point where the temperature exceeds 600 °F. Downstream of this point, ammonia can be partially removed in any particulate collection devices and could also be absorbed into any aqueous backend scrubbing systems. Ammonia released to the stack could potentially combine with hydrogen chloride to form a detached ammonium chloride plume. This phenomenon has been experienced with several ammonia-based, SNCR systems.[2] The urea-based, SNCR systems that have been on-going for more than 1 year have not experienced an ammonium chloride plume problem or any other backend NH_3 slip related problems.

The proportion of chemical used to reduce NO_x as compared to the theoretical reaction stoichiometry (chemical utilization) is about 50% at an NSR of 1. A very small portion (usually less than 5%) of the excess reagent shows up as NH_3 slip. The remaining excess reagent is converted to nitrogen, water, and carbon dioxide. This arises because oxidation of the NH_2 radical from the urea to NO_x competes with the NO_x reducing reaction.

Table 2

Location	Capacity	NO_x baseline
Mok-Dong Seoul, Korea	150 TPD	70–140 ppm @ 12% O_2
New Hanover County Wilmington, North Carolina	250 TPD	75–140 ppmvd
Bechtel SEMASS Rochester, Massachusetts	900 TPD	140–180 ppmvd @ 7% O_2

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[2] Howard B. Lange and Vikram Reddy, Carnot, Steven L. Dewitt, Biogen Power 1, Plume Visibility Related to Ammonia Injection for NO_x Control – A Case History; Presented at Council of Industrial Boiler Owners NO_x Control VII, Chicago, IL, 3 May 1994.

The application of SNCR has the benefit of smoothing the NO_x emissions from the normally erratic baseline conditions. Actual NO_x baseline ranges measured during several performance tests are listed in Table 2.

By characterizing NO_x formation vs. boiler load and making a feed-forward adjustment in reagent dosage, stack NO_x emissions are smoothed. Additional smoothing is accomplished by taking a feedback signal from the continuous emissions monitor and making a 'trim' reagent dosage adjustment.

SNCR is a post-combustion, add-on technology. As such, it can readily be combined with other technologies, such as reburning,[3] staged combustion,[4] or SCR.[5] The NO_x reduction performance of SNCR can easily be adjusted (or dialed up) by adjusting reagent feedrate. These abilities make SNCR ideal for application in a dynamic regulatory environment, and for use in an emissions averaging or trading plan.

Table 3
Application of NOxOUT to two muncipal waste combustors

Type of unit	MSWC	
Capacity (T/D)	360	720
Gross heat input (10^6 Btu/h)	150	300
NO_x baseline (ppmvd) (@ 7% O_2)	240	240
NO_x baseline (lbs/h)	62	124
Controlled NO_x (ppmvd) (@ 7% O_2)	120	120
NO_x reduction (%)	50	50
Capacity factor (%)	85	85
NO_x reduced (T/y)	115	230
Total erected cost for NOxOUT	*$537 000.00*	*$803 000.00*
Opertaing cost ($/Yr)		
O and M cost		
Power @ $0.04/kWh	5000	10 000
Chemical @ $0.85/gal[a]	53 600	107 594
Maintenance	11 000	11 000
Subtotal	*$69 860.00*	*$128 594.00*
Annualized capital cost[b]	61 755	92 310
First year cost, total	131 615	220 904
Cost per ton NO_x removed	*$1144.00*	*$960.00*

[a] Actual chemical costs to be obtained from licensed supplier.

[b] Fifteen-year life; 8% return on capital.

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[3] Hamid A. Abbasi and Mark J. Khinkis, Institute of Gas Technology, Richard Scherrer, Ogden Martin Systems, Inc., An Engineering and Economic Evaluation of the Methane de-NOx™ Technology.

[4] J.R. Comparato, Nalco Fuel Tech, R.A. Buchs, North American Chemical Corporation, and Dr. D.S. Arnold and L.K. Bailey, Kerr-McGee Corporation, NO_x Reduction at the Argus Plant Using the NOxOUT Process, Presented at the 1991 Joint Symposium on Stationary Combustion NO_x Control – EPA/EPRI, Washington, DC, 25–28 March 1991.

[5] Brian K. Gullett, US Environmental Protection Agency Air and Energy Engineering Research Laboratory, Paul W. Goff, Acurex Environmental Corporation, M. Linda Lin, Nalco Fuel Tech, James M. Chen, Pilot-Scale Testing of NO_x Removal with Combined Selective Catalytic Reduction and Selective Non-Catalytic Reduction, Presented at EPRI/EPA NO_x Symposium, Bel Harbour, FL, May 1993.

4. Project costs

The cost of applying a urea-based, NOxOUT SNCR system to typical 360 TPD and 720 TPD municipal waste incinerators is presented in Table 3. This covers the range of most incinerators. These costs are for application to a new facility. They exclude civil costs such as concrete pads for reagent storage, if required. For retrofits, these costs exclude boiler pressure part modifications which may or may not be required, and any business interruption costs associated with downtime for retrofit. It should be noted that the footprint for NOxOUT is extremely small and there is very little downtime, if any, required for a NOxOUT retrofit. These costs are effective for an installation in 1993.

5. Conclusion

SNCR has been proven around the world as an effective technology for reducing nitrogen oxide emissions from municipal waste incinerators. Typical NO_x reductions range from 50% to 80%. SNCR can be easily retrofitted to an existing municipal waste incinerator or designed into a new unit. SNCR's capital and operating costs fall well under most global air pollution cost guidelines for NO_x control. Potential process limitations such as by-product emissions can be controlled or eliminated by use of computer modelling techniques in designing the system. All of these factors will drive the increased regulatory prescription and use of SNCR technology on municipal waste incinerators.

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