

**SECOND GENERATION ADVANCED REBURNING
FOR HIGH EFFICIENCY NO_x CONTROL**

Quarterly Report No. 7 for Period

April 1 – June 30, 1999

Prepared by:

Vladimir M. Zamansky, Peter M. Maly and Vitali V. Lissianski

July 27, 1999

DOE Contract No. DE-AC22-95PC95251

Submitted by:

Energy and Environmental Research Corporation
18 Mason, Irvine, CA 92618

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Abstract

This project is designed to develop a family of novel NO_x control technologies, called Second Generation Advanced Reburning which has the potential to achieve 90+ NO_x control in coal fired boilers at a significantly lower cost than Selective Catalytic Reduction. The seventh reporting period in Phase II (April 1 – June 30, 1999) included experimental activities and combined chemistry-mixing modeling on advanced gas reburning. The goal of combustion tests was to determine the efficiency of advanced reburning using coal as the reburning fuel. Tests were conducted in Boiler Simulator Facility (BSF). Several coals were tested. The modeling effort was focussed on the description of N-agent injection along with overfire air. Modeling identified process parameters that can be used to optimize the AR-Lean process.

Table of Contents

| <u>Section</u> | | <u>Page</u> |
|--|--|-------------|
| Abstract | | 3 |
| Executive Summary | | 5 |
| 1.0 Coal Reburning Studies | | 6 |
| 1.1 Basic Coal Reburning Tests..... | | 6 |
| 1.2 Advanced Coal Reburning Tests..... | | 10 |
| 2.0 Kinetic Modeling | | 11 |
| 2.1 Model Setup | | 11 |
| 2.2 Model Validation..... | | 15 |
| 2.3 Parametric Study of the AR-Lean Process..... | | 17 |
| 2.4 Optimization of the AR Process..... | | 23 |
| 3.0 Future Work..... | | 24 |
| 4.0 Bibliography | | 24 |

List of Figures

| <u>Figure</u> | | <u>Page</u> |
|--|--|-------------|
| Figure 1-1. Basic coal reburning performance as a function of reburning heat input with natural gas primary | | 8 |
| Figure 1-2. Basic coal reburning performance as a function of reburning heat input with coal primary..... | | 9 |
| Figure 2-1. Reactor diagram of AR-Lean model setup..... | | 13 |
| Figure 2-2. Calculated temperature profile along centerline of the reburning jet .. | | 14 |
| Figure 2-3. Comparison of modeling predictions with experimental data..... | | 16 |
| Figure 2-4. Predicted effect of CO on NO _x reduction by urea | | 18 |
| Figure 2-5. Predicted effect of the reburning heat input on NO _x reduction | | 19 |
| Figure 2-6. Predicted effect of OFA/N-agent injection temperature | | 20 |
| Figure 2-7. Predicted effect of delay in N-agent injection on NO _x reduction | | 21 |
| Figure 2-8. Predicted effect of the amount of urea on NO _x reduction | | 22 |
| Figure 2-9. Predicted effect of the OFA/urea initial temperature..... | | 23 |

Executive Summary

This project is designed to develop a family of novel NO_x control technologies, called Second Generation Advanced Reburning which has the potential to achieve 90+ NO_x control in coal fired boilers at a significantly lower cost than SCR. The seventh reporting period in Phase II (April 1 – June 30, 1999) included combustion tests and combined chemistry-mixing modeling on advanced reburning.

The goal of combustion tests was to determine the efficiency of the advanced reburning (AR) process using coal as the reburning fuel. The first step in testing advanced coal reburning is to rank different coals in tests on basic reburning. Tests were conducted in 300 kW Boiler Simulator Facility (BSF). The fuel tested included coals utilized by South Carolina Electric and Gas (Consol Jines Fork, Sunnyridge, Knott-Floyt Land and Rocklick) and E-fuel, which is a waste-based product consisting of 70% coal, 15% plastics, and 15% paper. Test were conducted with natural gas and coals as the primary fuel. For the natural gas tests, E-Fuel provided the highest NO_x reduction. For the coal primary tests, Knott-Floyd Land coal performed the best. The data analysis to interpret the results is in progress.

The second step in advanced coal reburning testing includes combustion experiments with injection of N-agents in reburning and burnout zones, and along with overfire air (OFA). The tests matrix for these tests has been developed and tests are in progress.

Since the efficiency of AR depends on many factors, the best performance can be achieved if the effects of these factors on the process performance are well determined and understood. The most efficient approach to the AR optimization is to explore the effects of different parameters on NO_x reduction via combined kinetic-mixing modeling and to use the model for guidance to select the most effective test conditions. During the reporting period the extended AR model was developed and validated based on experiments involving co-injection of aqueous NH₃ and urea with OFA. The modeling effort during this period concentrated on parameters for optimizing the effect of N-agent addition with OFA, i.e. AR-Lean. The variables in modeling included the amount of the reburning fuel, temperature of flue gas at the point of OFA and N-agent injection, initial temperature of OFA and N-agent, and the amount of N-agent. Modeling shows that the efficiency of reburning increases when N-agent is co-injected with OFA. When optimum process parameters are selected, modeling predicts the efficiency of NO_x reduction in AR-Lean to be about 95%.

1.0 Coal Reburning Studies

Basic reburning and advanced reburning tests were performed in which coal was used as the reburning fuel. These tests were performed in conjunction with South Carolina Electric and Gas (SCE&G) that is considering installing basic coal reburning on multiple boilers with potential to subsequently utilize advanced coal reburning. Coal can be effectively used as a reburning fuel given the right fuel properties and process conditions. In some boiler applications the use of coal as a reburning fuel is limited by the fact that furnace temperatures and residence times are insufficient to fully combust the reburning coal, leading to high carbon-in-ash levels. However, the boilers of interest are equipped with carbon recovery units and thus can tolerate relatively high carbon-in-ash levels. Therefore, these boilers are ideal targets for application of coal reburning and advanced coal reburning.

Two test series were planned, including basic reburning and promoted advanced reburning. All test work was conducted at EER's 1.0 MMBtu/hr Boiler Simulator Facility, which was described in detail in previous reports. As detailed in the following sections, the basic reburning tests have been completed. The advanced reburning tests are in progress, the results will be reported in the forthcoming quarter and compare with the basic reburning tests.

1.1 Basic Coal Reburning Tests

Studies conducted by EER have shown that a number of fuels can be used effectively in the reburning process. However, due to the heterogeneous nature of coal, it is not possible to predict how a specific coal will perform as a reburning fuel based upon easily characterized fuel properties. Therefore, combustion tests were performed to evaluate the reburning performance of five coals of specific interest to SCE&G. The primary objective of the basic reburning tests was to characterize the impacts of reburning process parameters on NO_x reduction at conditions typical of the full-scale boilers.

For the initial experiments, the main burner was fired with natural gas. Ammonia was premixed with the combustion air to provide a controlled initial NO_x level. Five reburning test fuels were provided by the utility, including four bituminous coals and a material referred to as

“E-fuel,” which consists nominally of 70% coal, 15% plastics, and 15% paper. Fuel characteristics are shown in Table 1-1. Each fuel was pulverized such that 70% passed through a 200 mesh sieve. The coals were pulverized in a CE-Raymond deep bowl mill. E-Fuel was pulverized in a hammer mill. The E-Fuel was found to be somewhat sticky, especially at higher temperatures. It tended to clog the screen on the mill, requiring that the screen be removed and cleaned several times during the milling. It was also found during the reburning tests that the E-Fuel did not feed uniformly through the screw feeder.

Table 1-1. Test fuel analyses.

| Parameter | Units | Consol Jones Fork | Rocklick | Knott-Floyd Land | Sunnyridge | E-Fuel |
|------------------|--------|----------------------|----------|---------------------|------------|--------|
| Proximate | | | | | | |
| Moisture | % | 4.32 | 5.35 | 8.22 | 5.62 | 14.61 |
| Ash | % | 7.16 | 8.49 | 14.72 | 8.26 | 6.32 |
| Volatiles | % | 34.30 | 33.12 | 31.41 | 31.82 | 35.62 |
| Fixed Carbon | % | 54.22 | 53.04 | 45.65 | 54.3 | 43.45 |
| Total | % | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 |
| Calorific Value | Btu/lb | 13288 | 13217 | 11283 | 13124 | 11502 |
| Ultimate | | | | | | |
| Carbon | % dry | 78.63 | 78.77 | 69.50 | 77.03 | 75.07 |
| Hydrogen | % dry | 4.91 | 4.89 | 4.46 | 4.71 | 5.31 |
| Nitrogen | % dry | 1.62 | 1.50 | 1.39 | 1.41 | 1.60 |
| Sulfur | % dry | 0.82 | 0.91 | 1.11 | 0.82 | 0.78 |
| Ash | % dry | 7.48 | 8.97 | 16.04 | 8.75 | 7.40 |
| Oxygen | % dry | 6.54 | 4.96 | 7.50 | 7.28 | 9.84 |
| Total | % dry | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 |

The reburning fuel was injected into the furnace through an injector designed to provide rapid dispersion of the fuel into the flue gas. The transport medium for the fuel was nitrogen (to simulate recycled flue gas). The range of parameters investigated in the study represented the range of conditions available at the full-scale units. The main burner was fired at an excess air level of 10%. The reburning fuel was injected at a temperature of 2,600°F at rates between 10% and 20% of the total furnace heat input. For the tests with natural gas primary the OFA was

injected at 2100°F, corresponding to a reburning zone residence time of 800 ms. The initial NO_x level was set at 400 ppm on a dry, corrected to 3% O_2 basis.

Figure 1-1 compares reburning performance of the five fuels. The best performance was obtained with the E-Fuel, followed in order by Consol Jones Fork, Sunnyridge, Knott-Floyd Land, and Rocklick. It is believed that these trends are related to the composition and volatility of each reburning fuel. More volatile fuels tend to release the bound-nitrogen species and fuel fragments quicker. This allows the reburning chemistry more time to occur, and enables nitrogen-bound species to be processed in an environment where they can be reduced to molecular nitrogen.

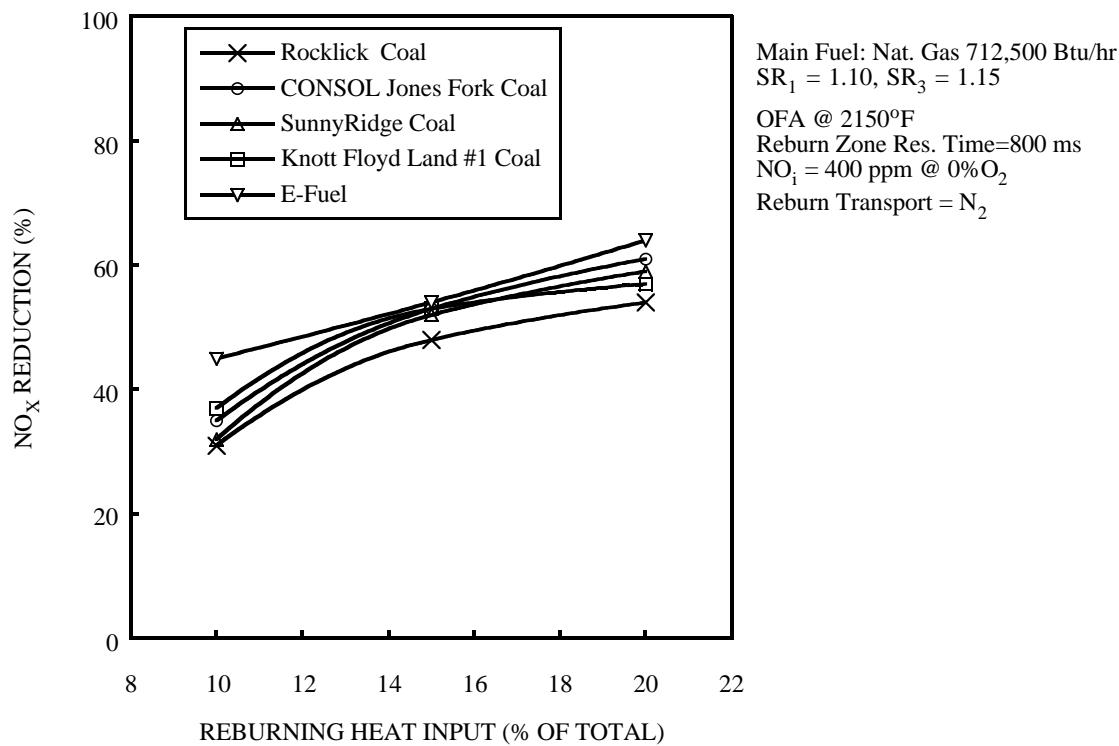


Figure 1-1. Basic coal reburning performance as a function of reburning heat input with natural gas primary.

Another factor that can affect reburning performance is the nitrogen content of the coal; higher nitrogen concentrations result in poorer reburning performance. However, in this case the nitrogen contents of the five test fuels vary by only 0.2%, so this factor has minimal impact on reburning performance.

The second series of tests was performed with coal as the primary fuel. For each of the four coals the same coal was used as both the main and reburning fuel. A limited amount of E-Fuel was available, so Sunnyridge coal was selected as the main fuel for the E-Fuel reburning tests. OFA was injected at 2300°F, corresponding to a reburning zone residence time of 400 ms. Figure 1-2 compares reburning performance of the five test fuels. For these conditions, Knott-Floyd Land performed best, followed by Rocklick, Consol Jones Fork, E-Fuel, and Sunnyridge. NO_x reductions for these tests were lower than those obtained during the previous tests with natural gas primary. This is mainly attributed to the fact that reburning zone residence time during the coal primary tests (400 ms) was lower than during the natural gas tests (800 ms).

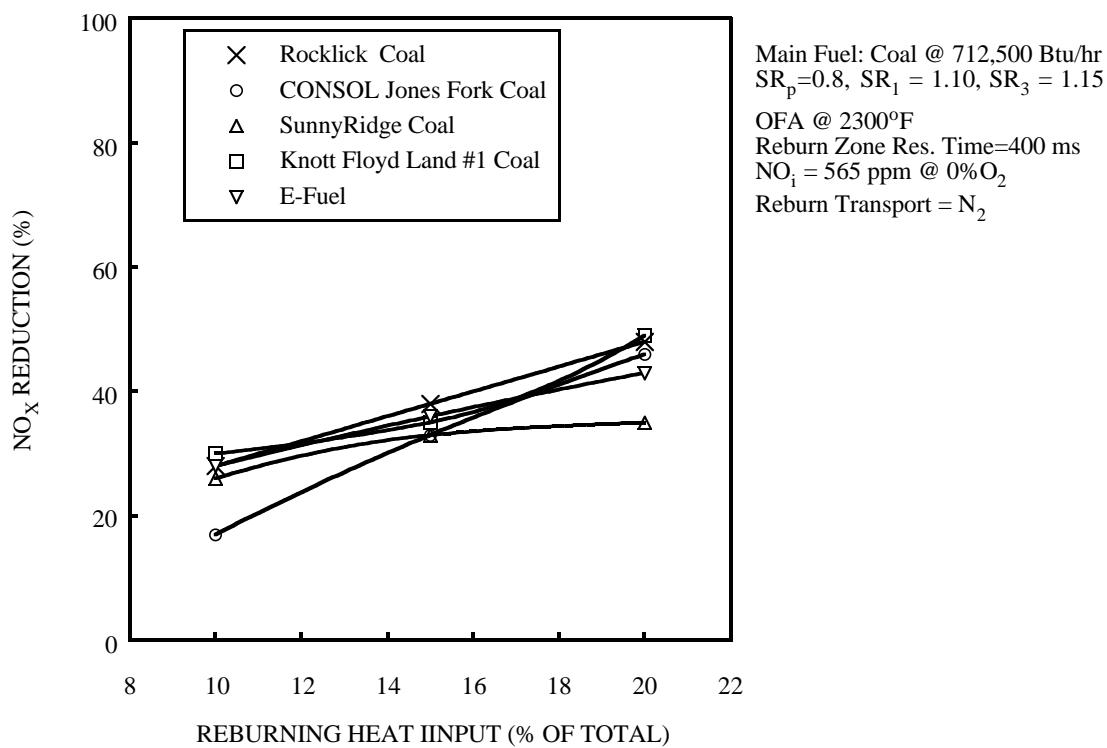


Figure 1-2. Basic coal reburning performance as a function of reburning heat input with coal primary.

In summary, the results of the experiments indicate that the four bituminous coals and E-Fuel would all be capable of providing reasonably high NO_x control at the conditions available at the full-scale boilers. At the BSF, at 20% reburning with nitrogen transport each of the five fuels provided over 50% NO_x reduction. Defining the level of control at full scale will depend on the extent to which effective mixing of the reburning fuel can be achieved, and the extent to which

the furnace flow field characteristics impact the reburning zone residence time. In addition, potential full-scale usage of E-Fuel would require that fuel pulverization and handling issues be addressed.

1.2 Advanced Coal Reburning Tests

The test approach involves performing basic reburning tests with the five fuels described above, followed by more detailed Advanced Reburning tests with the two fuels exhibiting best performance. Based on the basic reburning results, the Consol Jones Fork and Knott-Floyd Land coals were selected for AR tests. As summarized in Table 1-2, three test series will be performed, including AR-Lean, AR-Rich, and Reburning + SNCR. Natural gas will be used as the main fuel in initial test series followed by coal-over-coal tests. Variables will include reburning heat input, injection temperature of the OFA and additives, and promoter type and amount. After each variable is evaluated a series of optimization tests will be conducted. The objective will be to define maximum achievable NO_x control for advanced coal reburning. Performance will also be compared to that achievable with natural gas advanced reburning. These tests are in progress and will be reported in the upcoming quarter.

Table 1-2. Advanced coal reburning test matrix.

| Reburn Coal | Reburn % | OFA Temp., F | N-Agent | | Promoter | | |
|-----------------------|-------------|-----------------|---------|-----------|---------------------------------|--------|------------|
| | | | Type | Temp., F | Type | ppm | Location |
| 1. AR-Lean | | | | | | | |
| Consol Jones Fork | 10 | 1900-2300 | Urea | w/OFA | Na ₂ CO ₃ | 0-150 | w/N-agent |
| Knott-Floyd Land | 10 | 1900-2300 | Urea | w/OFA | Na ₂ CO ₃ | 0-150 | w/N-agent |
| 2. AR-Rich | | | | | | | |
| Consol Jones Fork | 10 | 1900 | Urea | 2000-2300 | Na ₂ CO ₃ | 0-150 | Reb. Zone |
| Knott-Floyd Land | 10 | 1900 | Urea | 2000-2300 | Na ₂ CO ₃ | 0-150 | Reb. Zone |
| 3. Reburn+SNCR | | | | | | | |
| Consol Jones Fork | 10-20 | 2300 | Urea | 1800-2000 | Na ₂ CO ₃ | 0-1200 | Downstream |
| Knott-Floyd Land | 10-20 | 2300 | Urea | 1800-2000 | Na ₂ CO ₃ | 0-1200 | Downstream |

2.0 Kinetic Modeling

This section describes ongoing efforts to model the AR process. Previous work¹ demonstrated that main features of AR can be described by using a detailed chemical mechanism² and with a one-dimensional representation of mixing. It was shown that injection of NH₃ and a sodium promoter in the reburning zone (AR-Rich) results in an increase of NO_x reduction. The modeling efforts during the reporting period concentrated on the effect of N-agent co-injection with OFA on NO_x reduction (AR-Lean).

During the reporting period the model¹ of basic reburning was expanded to describe the effect of N-agent co-injection with OFA on NO_x reduction (the AR-Lean process). The goal of this modeling effort is to predict the effect of AR conditions and optimize process performance.

2.1 Model Setup

Advanced reburning model was based on studies described in previous reports. The following features of the mixing process were addressed in modeling:

- Injected gases are available for the reaction over certain period of time (mixing time) rather than instantaneously.
- Injection of reburning fuel results in mixture stratification such that mixture composition in the mixing area is not uniform.

Since the characteristic time of chemical reactions in the reburning zone is usually smaller than that of the mixing process, distributed addition of reburning fuel and OFA can significantly affect the efficiency of the process. Mixture stratification in the reburning zone, or existence of local fuel-rich zones in which concentration of fuel is higher than average, is another important factor that affects efficiency of reburning. It is believed^{3, 4} that this factor is mainly responsible for high levels of NO_x reduction (40-50%) under slightly fuel-rich and even fuel-lean reburning conditions observed^{5, 6} in experiments.

The mixing process in the reburning zone was described in modeling by addition of flue gas to the stream of natural gas over mixing time (so-called inverse mixing). Additional fuel

stratification in modeling was introduced by assuming that within the reburning zone there is one zone with 25% larger and one zone with 25% smaller concentration of fuel than average. NO reduction was calculated as average of that for these two zones.

The current AR model is different from basic reburning model¹ in the following features:

- Model of inverse mixing was applied not only to the reburning zone, but to the injection of OFA and N-agent as well. Modeling shows that reactions in the burnout zone have small effect on NO_x concentration in basic reburning, except for large (more than 15%) heat inputs of the reburning fuel. Thus, choice of mixing mode in the burnout zone is not critical. In the AR-Lean process, significant NO_x reduction occurs upon injection of OFA and N-agent, and choice of the mixing mode is important.
- Model of a single jet in crossflow was used to calculate temperature profile within mixing zones. This was done to give more realistic representation of the temperature field in mixing areas.

The current model of AR-Lean treats the reburning process as series of four plug-flow reactors (Fig. 2-1). Each reactor describes one of the physical and chemical processes occurring in a boiler: addition of the reburning fuel, NO_x reduction as a result of the reaction with the reburning fuel, addition of OFA and N-agent, and NO_x reduction by N-agent and oxidation of partially oxidized products in the burnout zone. The first reactor describes mixing of the reburning fuel with flue gas. As was suggested earlier¹, the mixing zone was divided into two reactors *R1a* and *R1b*. The reactor *R1a* was assigned with more fuel-rich mixture than average, the reactor *R1b* with a less fuel-rich mixture. Averaging fuel contents distributed between the two reactors gave mixture composition corresponding to the total amount of added reburning fuel. The model of inverse mixing was applied to both reactors *R1a* and *R1b*. The mixture entering reactors *R1a* and *R1b* corresponds to natural gas. The mixture added to reactors *R1a* and *R1b* corresponds to products of natural gas combustion in air at SR₁ = 1.1 (main combustion zone). The flue gas was added to the reburning fuel with constant rate over 120 ms time. The second reactor *R2* described reactions in flue gas downstream from mixing area up to the point where OFA and N-agent were injected. The reburning fuel and flue gas were completely mixed in the second reactor. The third reactor *R3* described mixing of OFA and N-agent with flue gas

using model of inverse mixing. The mixture entering $R3$ consisted of air and N-agent. It was assumed that N-agent and OFA were premixed prior to injection. The gas added to $R3$ corresponds to products coming out of $R2$. The forth reactor $R4$ described reactions in the burnout zone.

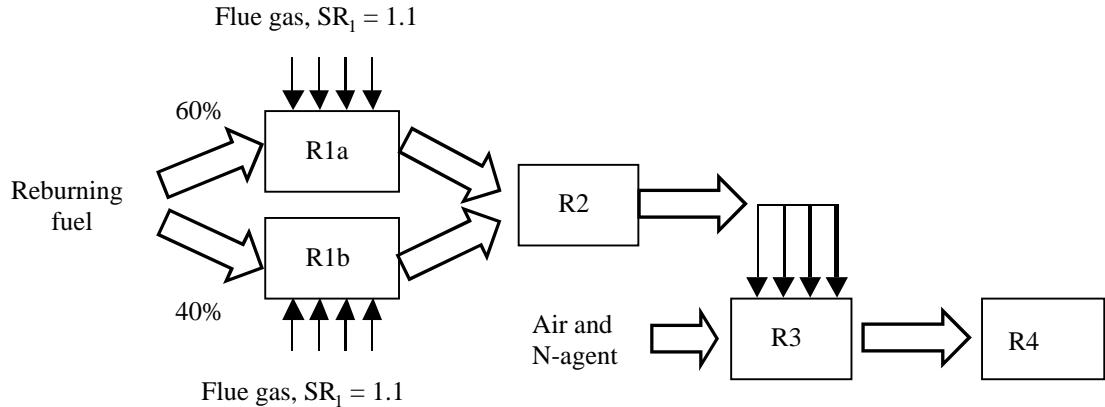


Figure 2-1. Reactor diagram of AR-Lean model setup.

Mixing time and temperature profile in mixing areas were estimated using a single jet in crossflow model, JICFIS⁷. Major inputs for the model included the velocity and density ratios of the crossflow to jet, their relative orientation in two dimensional rectangular coordinates, and the initial conditions (diameter, velocity, and temperature) of the jet. The mixing is determined by evaluating the entrainment rate of fluid from the crossflow into the jet. The JICFIS model was used up to the point where the amount of entrainment equals the main flux; after this, complete mixing is assumed. At the point of complete mixing the temperature of the jet is equal to that of the flue gas.

Figure 2-2 shows calculated temperature profile along the centerline of the reburning fuel jet for conditions used in experiments reported in previous reports. It was assumed in this calculations that reburning jet and flue gas consists of pure nitrogen. This approximation is not far from reality: flue gas consists of more than 70% nitrogen, and reburning fuel is co-injected with nitrogen to improve mixing with flue gas. Figure 2-2 shows that the mixing time of the reburning jet with flue gas is about 120 ms. By the end of the mixing time period, the temperature of the jet is 1670 K. It was assumed that variation in the amount of the reburning fuel has little effect on mixing time since reburning jet includes 70% nitrogen.

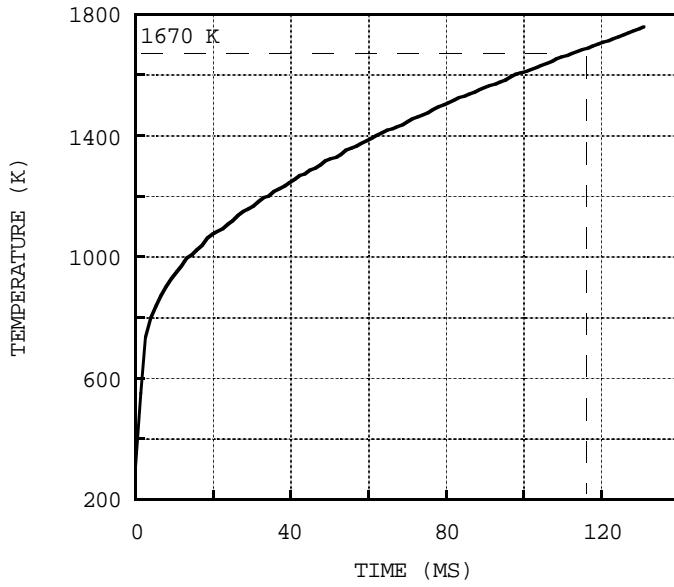


Figure 2-2. Calculated temperature profile along centerline of the reburning jet. Initial temperature of the reburning fuel is 300 K, temperature of flue gas is 1670 K.

OFA injection was handled in the same manner as the reburning fuel. For the OFA jet, the mixing time was calculated to be 110 ms. These times are approximate since the actual geometry of injectors does not precisely match the single jet treated by the JICFIS algorithm. However, the results are considered to reflect the magnitude of the mixing rate and approximate temperature profile in jet based on the general scale of the problem.

Based on the approximations in the JICFIS model, a single mixing time of 120 ms was used for all injections. Modeling shows that the value of the mixing time has strong effect on the predicted efficiency of the reburning process as mixing time increases from 0 (instantaneous mixing) to 100 ms. Further increase in mixing time has relatively small effect on NO_x reduction. Variation of mixing time within 120 ± 20 ms showed small effect on modeling results.

The kinetic model includes 447 reactions of 65 C-H-O-N chemical species (Glarborg et al.²). Initial amount $[\text{NO}]_i$ of NO was 600 ppm. Temperature of flue gas decreased at a linear rate -300 K/s . Variations in the temperature gradient within $\pm 50^\circ$ showed little effect on modeling predictions.

2.2 Model Validation

The EER chemical kinetic code ODF⁷, for “One Dimensional Flame was employed to model experimental data. ODF is designed to march through a series of well-stirred or plug flow reactors, solving a detailed chemical kinetics mechanism. ODF contains the same basic capabilities as Chemkin-II⁸, including the evaluation of pressure-dependent and reversible Arrhenius rate expressions, and the specification of time-dependent profiles of temperature and pressure. The solution algorithm has also been formulated to allow for the introduction of an arbitrary profile of heat and/or mass fluxes along the length of the reactor. This capability has proven significant for accurate modeling of many types of combustion systems, including reburning with natural gas.

The following parameters were varied in modeling:

- The amount of the reburning fuel (5-18% from the total amount of fuel).
- Temperature at which OFA and N-agent are injected (1200-1550 K).
- Type of the N-agent (NH₃ and urea).
- The initial temperature of OFA (300-600 K).
- Delay in injection of N-agent relative to OFA injection (0-0.6 s).
- The amount of the N-agent (NSR up to 3.0).

Previous work¹ showed that the model of basic reburning predicted main features of the reburning process. NO_x reduction efficiencies determined in experiments as functions of the initial NO_x amount, amount of the reburning fuel and OFA injection temperature are well-described by the model. During the reporting period the extended AR model was developed and validated based on experiments on co-injection of aqueous NH₃ and urea with OFA. The experimental data were obtained in a 300 kW Boiler Simulator Facility, described in previous reports and elsewhere¹.

Figure 2-3 shows comparison of modeling predictions with experimental data on the effect of OFA injection temperature in basic reburning and AR-Lean processes. In agreement with experiments, modeling predicts that the efficiency of the AR-Lean process increases as

temperature at which OFA and N-agent are injected decreases. Modeling predicts that urea is slightly more effective N-agent than aqueous NH_3 .

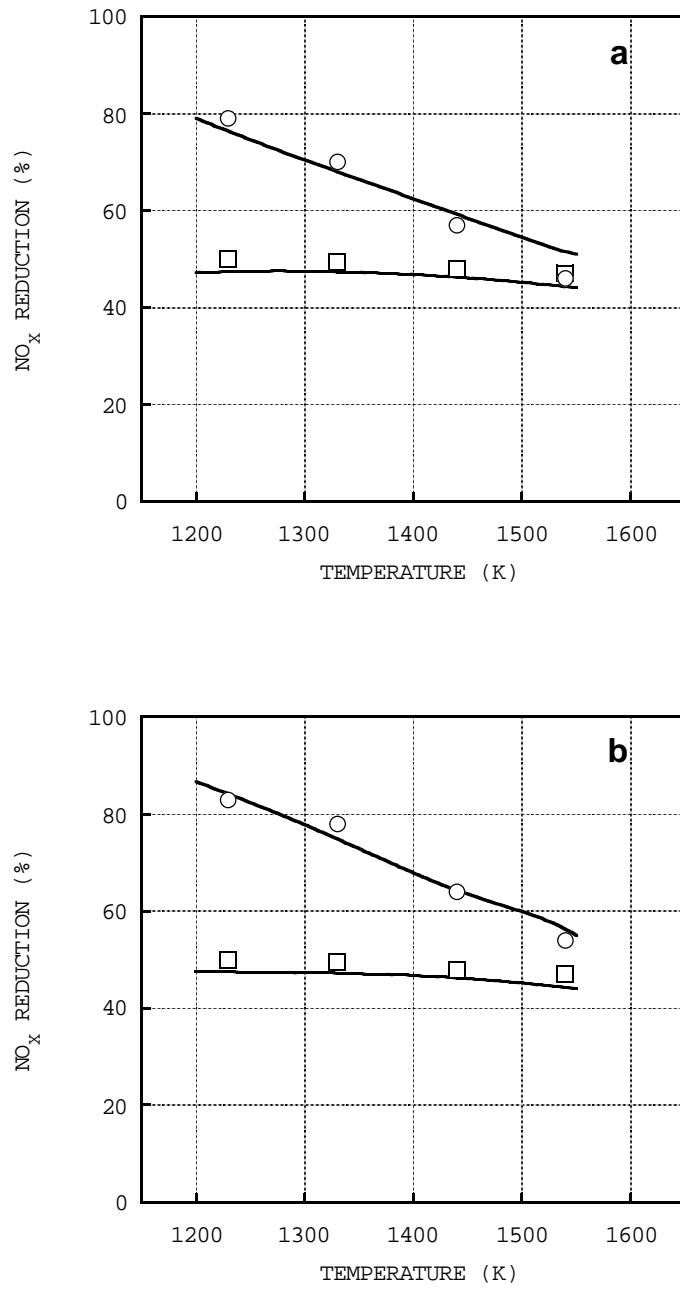


Figure 2-3. Comparison of modeling predictions (lines) with experimental data (symbols) on the effect of OFA injection temperature in AR-Lean with aqueous ammonia (a) and urea (b). Squares correspond to basic reburning, circles to co-injection of N-agent with OFA. NSR = 1.5. $[\text{NO}]_i = 600 \text{ ppm}$, 10% reburning.

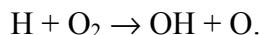
2.3 Parametric Study of the AR-Lean Process

Since the efficiency of AR depends on many factors, the best performance can be achieved if the effects of these factors on the process performance are well determined and understood. The most efficient approach to the AR optimization is to explore the effects of different parameters on NO_x reduction via kinetic modeling, and use model for guidance to select the most effective test conditions. The following sections describe results of modeling study on the effect of the AR-Lean parameters on NO_x reduction.

Effect of CO on NO_x Reduction

Fuel fragments, such as CO, formed in the reburning zone can significantly affect NO_x reduction in the presence of N-agents⁹. However, previously it was difficult to determine this effect quantitatively since a predictive model of AR-Lean had not been developed. The described model of AR-Lean has such quantitative predictive capabilities.

Figure 2-4 shows the predicted effect of CO on NO_x reduction by urea injection. Urea and air were injected into flue gas containing 600 ppm NO, 8% CO_2 , 16% H_2O , balance N_2 (corresponds to typical composition of flue gas entering the burnout zone). The amount of CO in flue gas was varied in modeling from 0 to 3%. Marks “10%” and “18%” on x -axis indicate the amount of CO coming from the reburning zone at 10 and 18% reburning fuel. At 1280 K the presence of CO in flue gas promotes NO_x reduction, while at 1530 K NO_x reduction is inhibited by CO. The relative effect of CO at 1280 K also depends on the amount of CO: small amounts of CO significantly promote NO_x reduction, while CO concentrations on the level 3% have much smaller effect than that expected from linear interpolation. Thus, CO can increase or decrease the efficiency of the AR-Lean process depending on the temperature at which OFA and N-agent are injected, and on CO concentration. At high temperatures, which are characterized by high concentrations of active species in flue gas, the CO oxidation reduces the efficiency of N-agent by competing for radicals with reactions of NO_x reduction. At low temperatures, the concentrations of active species in flue gas are much smaller, and the CO oxidation produces radicals via chain reaction



The similar observation was made by Leckner et al.¹⁰ for the effect of CO on Selective Non-Catalytic Process.

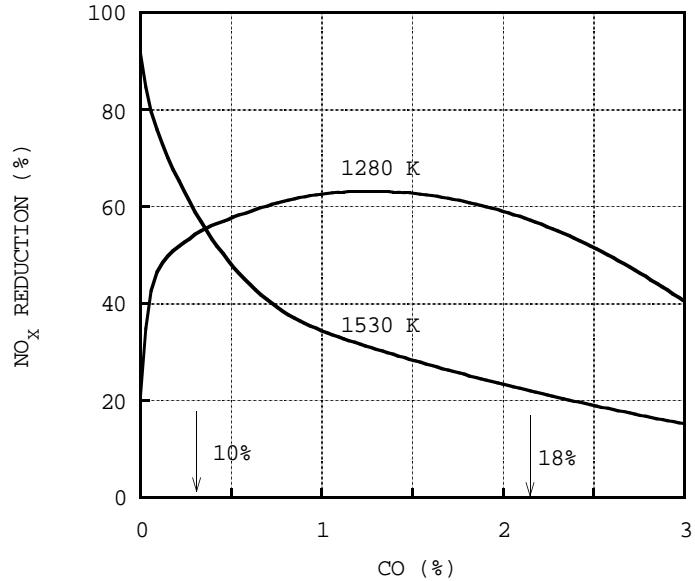


Figure 2-4. Predicted effect of CO on NO_x reduction by urea. $[\text{NO}]_i = 500 \text{ ppm}$, NSR = 1.8.

Effect of Reburning Heat Input

Since the amount of CO present in flue gas at the point of OFA injection depends strongly on the amount of fuel injected in the reburning zone, the efficiency of the N-agent also depends on heat input of the reburning fuel. In basic reburning, efficiency of NO_x reduction increases as the amount of the reburning fuel increases. It also slightly increases as OFA injection temperature decreases (Fig. 2-3). The situation is quite different when N-agent is co-injected with OFA (Fig. 2-5).

At OFA and N-agent injection temperatures in the range of 1280-1530 K, AR-Lean is more effective than basic reburning for heat inputs of the reburning fuel less than 13%. This is because the amount of CO entering the burnout zone is relatively small (less than 5,000 ppm) for reburning heat inputs less than 13%. Modeling predicts that the concentration of CO in flue gas at 18% heat input of the reburning fuel is approximately 2.3% At 1530 K, this amount of CO has strong inhibiting effect on NO_x reduction. At 1280 K, the promotion effect of CO is relatively

small. High concentrations of H_2 in flue gas coming from the reburning zone also contribute to the degradation of AR-Lean performance at large heat inputs of the reburning fuel. Modeling suggests that the AR-Lean process is more effective than basic reburning at heat inputs of the reburning fuel smaller than that usually utilized in basic reburning. Figure 2-6 supports these modeling conclusions by demonstrating that AR-Lean with 5 and 10% reburning is more effective than 18% reburning at all reasonable OFA injection temperatures. Note, however, that these modeling results are obtained under assumption that N-agent and OFA are perfectly mixed prior to injection. The effect of the delayed N-agent injection will be discussed in the following section.

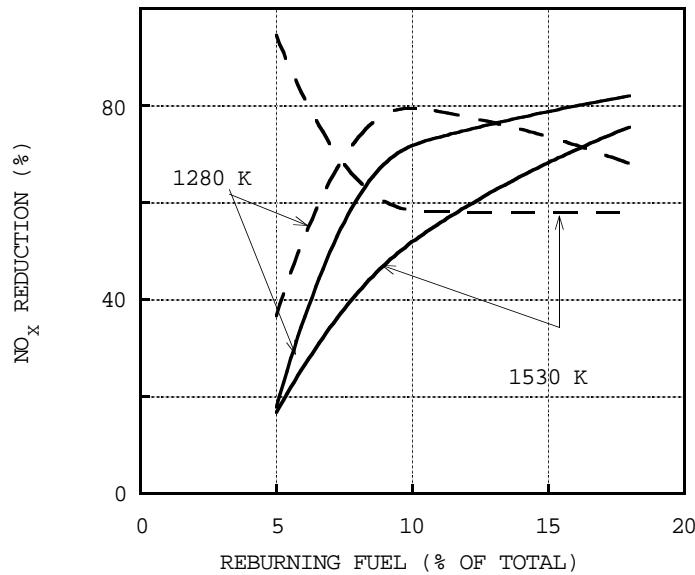


Figure 2-5. Predicted effect of the reburning heat input on NO_x reduction at different OFA injection temperatures. Solid lines correspond to basic reburning, dashed lines to AR-Lean.

Thus, the AR-Lean process not only provides higher levels of NO_x reduction than basic reburning, but it also requires less reburning fuel. Modeling suggests that the efficiency of the AR-Lean process depends on the amount of the reburning fuel and OFA injection temperature. The amount of the reburning fuel determines the composition of flue gas entering the burnout zone. Among other species present in flue gas, CO has the strongest effect on the efficiency of N-agent. The larger the reburning heat input, the more CO is present in flue gas by the end of the

reburning zone. Figure 2-5 shows that for 18% reburning, the presence of CO in flue gas decreases the efficiency of N-agent at OFA injection temperatures in the range of 1400-1500 K.

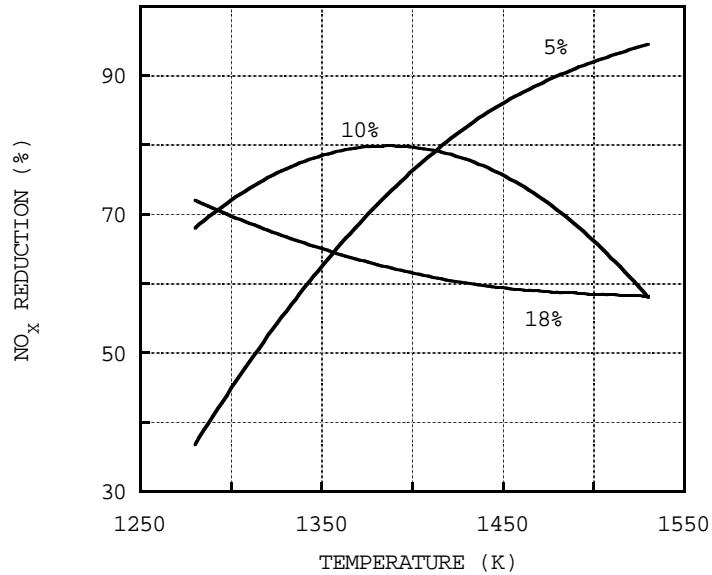


Figure 2-6. Predicted effect of OFA/N-agent injection temperature.

Effect of N-Agent Injection Delay Time

To eliminate negative influence of CO on NO_x reduction, the N-agent can be injected with a delay, or injection can be arranged in such a way that the release of N-agent into the gas phase occurs with a delay. The later can be done, for example, by injection of large droplets of aqueous solution containing N-agent. Because of the time required for droplets to evaporate and mix with flue gas, the N-agent will be delivered to the flue gas with some delay. Both approaches result in N-agent entering flue gas after the mixing process of OFA and flue gas is almost completed and thus allow for most of the CO to be oxidized before N-agent reacts with NO_x.

Figure 2-7 shows the predicted effect of urea injection delay time on NO_x reduction in AR-Lean process at OFA injection temperature 1530 K. As the delay time increases, the efficiency of the process also increases. At long delay times the dependence becomes less prominent since at 1530 K most CO is oxidized within first 200 ms.

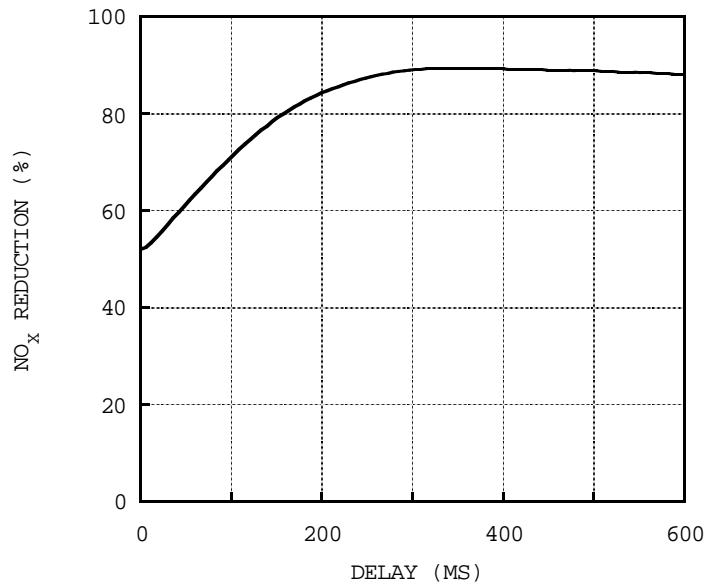


Figure 2-7. Predicted effect of delay in N-agent injection on NO_x reduction. 10% reburning, [NO]_i = 600 ppm, OFA is injected at 1530 K.

Effect of the Amount of N-Agent

Figure 2-8 shows the effect of the amount of urea on NO_x reduction at different OFA/N-agent injection temperatures calculated for zero delay times of N-agent injection. There is an optimum amount of N-agent that results in the highest efficiency of the process at OFA/N-agent injection temperatures in the range 1430-1530 K. As OFA injection temperature decreases, optimum becomes less prominent. Modeling shows that for zero delay times and OFA injection temperatures of 1430 and 1530 K, the AR-Lean process is most effective for NSR equals 1.5 and 1.25, respectively.

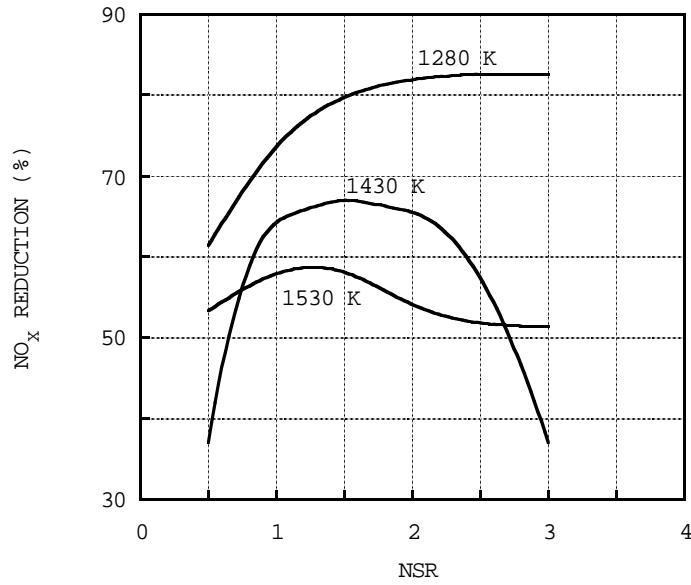


Figure 2-8. Predicted effect of the amount of urea on NO_x reduction. 10% reburning.

Effect of the Initial OFA/N-Agent Temperature

Figure 2-9 shows the effect of the initial OFA/N-agent temperature on NO_x reduction. Initial OFA/N-agent temperature is defined here as temperature of OFA/N-agent mixture prior to injection into flue gas. Preheating OFA results in decrease of NO_x reduction efficiency. This effect does not depend on the temperature of flue gas at the point of OFA injection. The main reason for the negative impact of elevated initial temperatures of OFA on NO_x reduction is that the increase in OFA initial temperature results in decrease of the mixing time of OFA and flue gas and as mixing time in burnout zone decreases, the efficiency of the process also decreases. For example, modeling shows that as the initial temperature of OFA increases from 300 K to 600 K, the mixing time decreases from 120 ms to 80 ms and NO_x reduction decreases on average by ten percentage points.

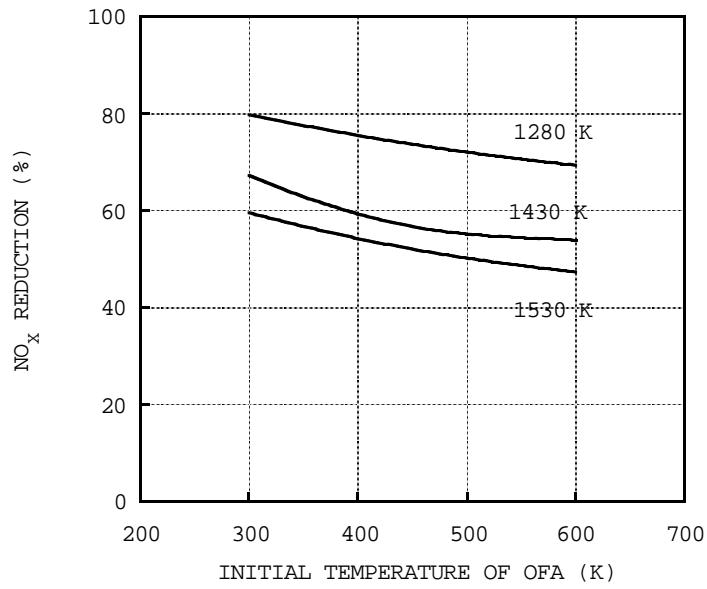


Figure 2-9. Predicted effect of the OFA/urea initial temperature on NO_x reduction at 10% reburning.

2.4 Optimization of the AR Process

Modeling demonstrated that several process parameters could be used for optimizing AR-Lean. The following parameters were considered:

- The amount of the reburning fuel.
- Temperature of flue gas at the point of OFA/N-agent injection.
- The delay in N-agent injection (separate injection or large droplets).
- The amount of N-agent.
- The initial temperature of OFA/N-agent.

Modeling suggests that selection of these parameters in the optimum range results in the efficiency of the AR-Lean process over 90%.

Assuming that the amount of N-agent is in the optimum range identified by modeling (for example, $\text{NSR} = 1.5$), the following conditions result in the highest NO_x reduction: the amount of the reburning fuel is in the range of 5-10%, the OFA injection temperature is in the range of 1350-1500 K. The AR-Lean process is more efficient at lower heat inputs of the reburning fuel

and higher OFA injection temperatures. The efficiency of AR-Lean can be further increased if the N-agent is injected with a delay. Optimization of AR via modeling is still in progress.

3.0 Future Work

This report describes ongoing efforts to optimize the AR process. Future test activities will include experimental evaluation of advanced coal reburning. Kinetic modeling on optimization of AR variants will continue. The effect of additives (N-agents and promoters) will be predicted using the model and compared with experimental data.

4.0 Bibliography

1. Zamansky, V.M. and Lissianski, V.V., *Israel Journal of Chemistry*, Special Issue: Combustion; Effect of Mixing on Natural Gas Reburning, 39:63-71 (1999).
2. Glarborg, P., Alzueta, M.U., Dam-Johansen, K., and Miller, J.A. *Combust. Flame* 115:1 (1998).
3. Luan, T., Liu, H., Gibbs, B.M., and Hampartsoumian, E. (1996) *The 9th International Symposium on Transport Phenomena in Thermal-Fluids Engineering*, p. 268.
4. Miller, C.A., Touati, A.D., Becker, J., and Wendt, J.O.L. (1998) *27th Symposium (International) on Combustion*, Abstracts of Accepted Papers, p. 122, The Combustion Institute, Pittsburgh.
5. Kokkinos, A. *EPRI Journal* 8, 17:36 (1992).
6. Folsom, B.A., Sommer, T.M., and Royne. (1991) *AFRC-JFRC International Conference on Environmental Control of Combustion Process*, Paper 16.
7. Kau, C.J., and Tyson, T.J. (1987) A Computer Program for General Flame Analysis. *U.S. Environmental Protection Agency Report No. EPA-6000/7-87-027*.
8. Kee, R.J., Rupley, F.M. and Miller, J.A., *Chemkin II: a Fortran Chemical Kinetics Package for the Analysis of Gas Phase Chemical Kinetics*, Sandia National Laboratories Report SAND89-8009, 1992.

9. Zamansky, V.M., Ho, L., Maly, P.M., and Seeker, W.R., *The Fourth International Conference on Technologies and Combustion for a Clean Environment*, Lisbon, Portugal, 1997.
10. Leckner, B., Karlsson, M., Kilpinen, P., and Hupa, M., *Ind. Eng. Chem. Res.* 30:2396-2404 (1991).