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An Experimental Study of NO_x Recycle in the NOXSO Flue Gas Cleanup Process

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1.0 EXECUTIVE SUMMARY

In the previous reporting period (from January to March), efforts focused on solving the problems encountered during shakedown tests, and modifying the gas injection and measurement system to ensure the accuracy of test data. After the system was modified, the mass balance between the NO amount injected into the combustion air duct and the NO_x measured at the stack was checked without combustion. Most of the tests showed a very good NO_x mass balance (between 92% to 95%).

The tests of NO injection with combustion have demonstrated very promising results. The test data showed quite consistent NO_x reduction efficiencies, which is similar to the previous test results on the pulverized coal combustor at DOE-PETC. An average of about 70% NO_x destruction efficiency was achieved for the NO injection into the primary air duct independent of injected NO flow rates and exit O₂ concentrations. About 58%, 50%, and 46% average destruction efficiencies were obtained for the NO injection into the secondary air duct corresponding to the exit O₂ concentrations of 2%, 3%, and 4% respectively.

2.0 INTRODUCTION

NO_x recycle is one part of the NOXSO process. In this process, 90% of the acid pollutants (NO_x and SO_2) can be removed simultaneously by adsorption on the surface of a sorbent material. The sorbent is subsequently regenerated by heating and contacting the hot sorbent with a reducing gas followed by steam. The NO_x is removed in the heating process, and the SO_2 is removed by the chemical reaction of the reducing gas and steam. The NO_x stream produced is returned to the combustion process with combustion air, which is called the NO_x recycle. The NO_x is reduced in the flame to N_2 , H_2O , and CO_2 .

The NO_x recycle concept operates by means of two different mechanisms. The first relies on the thermodynamic and kinetic conditions in the flame. When NO_x is fed to the coal combustor at concentrations greater than the equilibrium concentration, a portion of the recycled NO_x is reduced to N_2 and O_2 to approach the equilibrium concentrations (from high to equilibrium value). The kinetic rate of NO formation also decreases due to the high initial recycled NO concentration. The second mechanism relies on the reducing nature of the combustor flame. Free radicals generated at flame temperatures react with the NO_x species reducing them to N_2 and H_2O or CO_2 . Therefore, reaction equilibrium and the reducing atmosphere in the flame both serve to destroy recycled NO_x species.

NO_x recycle will be implemented at the full-scale commercial demonstration plant at Niles. The concept of NO_x recycle has been tested previously at DOE-Pittsburgh Energy Technology Center (PETC) using a 500 lb/hr coal combustor used for the 3/4 MW tests and also using a tunnel furnace capable of being fired with a variety of fuels including gas, fuel oil, coal, and coal-water mixtures. The results were very promising and demonstrated the destruction of NO_x compounds when fed to the combustion system. The data also indicate that the extent of NO_x reduction is determined by the location of the NO_x injection and the combustor design.

This current project is pilot-scale testing of NO_x recycle. The objectives are to provide further fundamental understanding of the NO_x destruction mechanisms and to help with the

implementation of NO_x recycle at the Niles station. A Babcock & Wilcox 6 million Btu/hr Small Boiler Simulator (SBS) (see Figures 1 and 2) was chosen to perform the tests. The SBS is the same type of cyclone furnace as that at Niles and simulates a large cyclone unit very well (see Table 1).

The project involves conducting a series of tests which include injecting NO, NO₂, N₂O, and a simulated NO_x recycle stream respectively. The work performed during this reporting period focused on completing NO injection testing series, and conducting NO simultaneous injection and NO₂ injection testing series. The NO injection test data at lower furnace load (4MBTU/HR) still showed quite consistent NO_x reduction efficiencies, which is similar to that at full load (6MBTU/HR) condition and the previous test results on the pulverized coal combustor at DOE-PETC. The NO simultaneous injection test data showed the higher percentage of NO injected into the primary air duct, the better the NO_x reduction efficiency. The NO₂ injection test results demonstrated a little higher NO_x reduction efficiencies (average about 67% when injecting NO₂ into the secondary air duct)) than that in the NO injection tests with the same conditions.

3.0 PROJECT DESCRIPTION

In order to provide further fundamental understanding and practical information for NO_x recycle implementation at the Niles station, the following four tasks have been defined to be performed in this project:

Task 1: Project management and reporting. This includes overall management of the project, preparation of monthly reports, preparation of draft and final project work plan, supervision of tests performed by B&W, and final report preparation.

Task 2: Experimental Testing. This has been subcontracted to B&W. It involves site preparation and testing. A schematic diagram of the NO injection and sampling system is shown in Figure 3. Pure NO gas from the gas cylinder goes through a rotameter and is injected into

the combustion air duct a short distance from the furnace. The sampling probes for the inlet NO and NO_x measurement are installed downstream of the injection location and before the cyclone. Both inlet and stack NO and NO_x concentrations are measured by continuous NO/NO_x analyzers. The tests consist of shakedown, baseline, simulated NO recycle, simulated NO₂/N₂O recycle, and novel concept tests. The NO, NO₂ and N₂O will be injected separately to investigate their effects on the NO_x destruction in the furnace. The injection of a simulated NO_x recycle stream will also be tested. The injection location will be in the primary air alone, secondary air alone, and both primary air and secondary air together. The parameters varied during the tests are furnace load, outlet oxygen concentration, and the amount of injected gases which are multiples of the baseline NO_x production levels.

Task 3: Data Analysis. The collected data will be analyzed. Data will be compiled into tables and graphs. The test data will be compared with previous NO_x recycle test results using pulverized coal combustor (performed at PETC) and recommendations for burner operation and NO_x recycle will be prepared.

Task 4: Comparison of B&W's pilot scale boiler to Ohio Edison's commercial boiler at Niles, Ohio. The end result of this task will be a comparison of the two units and suggested conditions for the NO_x recycle tests.

4.0 PROJECT STATUS

Experimental Testing

Management and Reporting: A site visit was made by Mr. Tony Mayne of the Department of Energy (DOE) - Pittsburgh Technology Energy Center (PETC) to the B&W Alliance Research Center. The test matrix, schedule, financial status, and some of the problems that occurred during testing were discussed. A tour of the facility was given while testing was in process.

Several meetings were held between NOXSO and B&W, and between DOE-PETC and NOXSO to discuss the final test matrix, time extension and additional funding for the completion of the project. Decision has been made by NOXSO and approved by Ohio Edison and W. R. Grace to cancel the N₂O testing series, because there was no N₂O found in the NO_x recycle stream in the pilot plant tests of NOXSO Process at Toronto, OH.

Due to the delay of the test program at B&W, DOE has approved the extension of the project. The final report will be due by October 31, 1992.

B&W has requested additional funding to complete the experimental test program. NOXSO has submitted a proposal to DOE-Pittsburgh Energy Technology Center which includes the final test matrix and additional cost estimation.

Site Preparation: Site preparation activities were completed on the nitrogen dioxide injection system. The lines were pressure and leak checked and approved by the Quality Assurance Group of B&W. After the shakedown test of NO₂ injection, the NO₂ injection system was further modified. The methane injection system was also installed.

Testing: NO injection tests have been completed. Total 48 tests of NO injection were conducted with four NO injection flow rates, three exit O₂ concentrations (2%, 3%, and 4%), two injection locations (primary air duct and secondary air duct) and two furnace loads (6 and 4 MBTU/HR). NO injections at full load (6MBTU/HR) conditions were tested in the last reporting period. NO injections at lower load (4MBTU/HR) conditions were tested in the present reporting period. The test results showed no effect of furnace load on the NO_x reduction. Therefore, it is not necessary to perform tests at furnace load of 5MBTU/HR condition as planned in the original test matrix.

A few simultaneous NO injection tests were conducted. In these tests, NO gas was injected into both primary air duct and secondary air duct together. The test conditions are

furnace load of 6MBTU/HR and Exit O₂ concentration of 2%. In order to simulate the operating condition at Niles plant, the ratio of NO gas injected into primary air duct to that injected into secondary air duct was 15/85 in most tests. This ratio was also varied to investigate the effect of injection location.

Shakedown tests of NO₂ injection have been done to check the injection system. After modifying the system, a few tests were conducted to inject NO₂ into the secondary air duct.

The procedures for each test were:

- 1). Measure the baseline stack NO_x before injection of NO.
- 2). Inject NO or NO₂ and take data at the steady state condition.
- 3). Stop injection and measure the baseline condition again before next injection.

Preliminary data analysis of the completed tests is given in the following section.

Data Analysis

The preliminary test data for the completed NO and NO₂ injection tests are shown in Figures 4 - 10. In these figures, the NO_x reduction efficiency is defined as follows:

$$eff. \% = \frac{NO_{xi} + NO_{xb} - NO_{xs}}{NO_{xi}} * 100 \quad (2)$$

where, NO_{xi} is the feed rate of pure NO_x gas (lb/hr), NO_{xb} is the baseline stack NO_x without NO_x injection (lb/hr), and NO_{xs} is the stack NO_x with NO_x injection (lb/hr).

F* is the normalized NO feed rate which is given by:

$$F^* = \frac{NO_{xi}}{NO_{xb}} \quad (3)$$

The normalized NO_x production rate is calculated according to the following equation:

$$R^* = \frac{R}{R_0} = \frac{NO_{xs} - NO_{xi}}{NO_{xb}}$$

where, R is the net NO_x production rate which equals the NO_x flow rate at the exit of the combustor minus the NO feed rate into the combustor; R_0 is the NO_x production rate at $NO_{xi}=0$, i.e. the baseline NO_x flow rate at the exit of the combustor. The figures, which show the normalized NO_x production rate (R^*) vs. normalized NO_x feed rate (F^*), also indicate the NO_x reduction efficiency if the data can be represented by the straight line described by the equation $R^*=1-aF^*$. It can be derived that the parameter "a" is the slope of the line and represents the NO_x reduction efficiency (eff. %).

In the present tests performed at B&W, when pure NO is injected into the primary air duct of the cyclone furnace (see Figures 4 and 5), the test data show quite consistent NO_x reduction efficiencies. The furnace loads are 6 MBTU/HR in the tests of Figure 4, and 4 MBTU/HR in the tests of Figure 5. From these figures, there is no effect of furnace load on the NO_x reduction efficiency. The average NO_x reduction efficiency is about 70%, which is also not affected by the exit O_2 concentrations. This is quite similar to the test results on the pulverized coal combustor at DOE/PETC, as shown in Figure 11, which shows a constant NO_x reduction efficiency of 65% for the pure NO injection into the primary air duct.

When pure NO is injected into the secondary air duct of the cyclone furnace (see Figures 6 and 7), the NO_x reduction efficiency is lower than that of the NO injection into the primary air duct and slightly varies with the exit O_2 concentrations. At the condition of 6 MBTU/HR furnace load, the average reduction efficiencies are about 58%, 50% and 46% corresponding to

the exit O₂ concentrations of 2%, 3%, and 4% respectively. However, at the condition of 4 MBTU/HR furnace load condition, there is little difference in the NO_x reduction when exit O₂ concentration varies from 3% to 4%. The lower reduction efficiency and the slight effect of the exit O₂ concentrations for the secondary air duct injection are attributed to the less reducing atmosphere in the secondary air region.

Figures 8 and 9 show the results of NO simultaneous injection tests. In the tests of figure 8, about 15% of total NO is injected into primary air duct and about 85% of total NO is injected into secondary air duct. The test data showed the same trend as other tests. About 56% NO_x reduction efficiency was indicated, which is close to the results of NO injection into the secondary air duct alone. Figure 9 displays the NO_x reduction efficiency vs. percentage of total NO injected into primary air duct. As expected, the more NO injected into the primary air duct, the higher the NO_x reduction efficiency.

The test results of NO₂ injection into the secondary air duct are shown in Figure 10. The furnace load is 6 MBTU/HR in these tests. Consistent NO_x reduction efficiencies were also demonstrated. Average NO_x reduction efficiency is about 67%, which is higher than that of NO injection at the same furnace conditions. It should be noted that the test data shown in Figures 4-10 are preliminary. Further data reduction is needed for the final evaluation.

5.0 PLANNED ACTIVITIES

Testing will continue to complete the tests of NO simultaneous injection series, NO₂ injection series, addition of methane injection and simulated mixture testing defined in the work scope. Data reduction and analysis will be performed. Final report will be written and submitted to DOE-PETC by October 31, 1992.

6.0 SUMMARY

During the current reporting period, NO injection series of tests series have been completed. Part of NO simultaneous injection and NO₂ injection tests were conducted. All the tests results showed consistent trends. About 70% average NO_x reduction efficiency has been achieved when NO is injected into the primary air duct, which was not affected by the NO injection flow rate, the furnace load and exit O₂ concentration. When NO is injected into the secondary air duct, the NO_x reduction efficiency ranges from 46% to 60% with dependence on the exit O₂ concentration. The NO simultaneous injection tests showed that the NO_x reduction efficiency increases with the percentage of total NO injected into the primary air duct. About 67% average NO_x reduction efficiency was demonstrated with injection of NO₂ into the secondary air duct, which is higher than that with injection of NO at the same test conditions.

Table 1. Comparison of Baseline Conditions Between the SBS Facility and Commercial Units		
	SBS	Typical Cyclone-Fired Boilers
Cyclone Temperature	> 3000°F	> 3000°F
Resident Time	1.4 sec. at full load	0.7-2 sec.
Furnace Exit Gas Temperature	2265°F	2200°-2350°F
NO _x Level	900-1200 PPM	600-1400 PPM
Ash Retention	80-85 %	60-80 %
Unburned Carbon	< 1% in Ash	1-20 %
Ash Particle Size (MMD; Bahco)	6-8 microns	6-11 microns

Figure 1 Small Boiler Simulator (SBS) Schematic

Figure 2 Cyclone Furnace

Schematic of NO Injection and Sampling System

Figure 3

**PRELIMINARY DATA OF NO_x RECYCLE
ON B&W'S CYCLONE FURNACE
(NO Injection into the Primary Air Duct)**

Figure 4

**PRELIMINARY DATA OF NO_x RECYCLE
ON B&W'S CYCLONE FURNACE
(NO Injection into the Primary Air Duct)**

Figure 5

**PRELIMINARY DATA OF NO_x RECYCLE
ON B&W'S CYCLONE FURNACE
(NO Injection into the Secondary Air Duct)**

Figure 6

**PRELIMINARY DATA OF NO_x RECYCLE
ON B&W'S CYCLONE FURNACE
(NO Injection into the Secondary Air Duct)**

Figure 7

**PRELIMINARY DATA OF NO_x RECYCLE
ON B&W'S CYCLONE FURNACE
(NO Injection into PA & SA Ducts Simultaneously)**

Figure 8

**PRELIMINARY DATA OF NOx RECYCLE
ON B&W'S CYCLONE FURNACE**
(NO Injection into PA & SA Ducts Simultaneously)

Figure 9

NO2 INJECTION INTO THE SECONDARY AIR DUCT

Figure 10

EXPERIMENTAL DATA OF NOx RECYCLE ON PETC'S PC COMBUSTOR (Pure NO Injection into the Primary Air Pipe)

Figure 11