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**Computational Modeling and Experimental Studies on NO_x Reduction
Under Pulverized Coal Combustion Conditions**

Technical Progress Report
Fifth Quarter

January 1, 1996 - March 31, 1996

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INTRODUCTION

During this quarter (January-March 1996), the focus was on getting the experimental facility ready for operation. Upon receiving all the pieces of equipment, the research team, first spent a great deal of time in calibration of various instruments and then, began assembling the pieces for the experimental work. The use of Swage-lok fittings helped in arresting the gas leaks fairly easily. The entire assembly consisted of teflon tube connections a) from gas cylinders to the flow meters, manifold and buffer vessel, b) from buffer vessel to the inlet of the reactor enclosed in the middle, by a furnace, c) from the exhaust end of the reactor to the NO_x analyzer via gas dryer and d) from the analyzer to the vent. Details of the experimental setup are given in the subsequent section.

Once the setup was checked for leaks and the leaks were arrested, the facility was operational. Several experiments on NO reburning with methane were undertaken. The flow rates for various gases were calculated for five reburning stoichiometric ratios and the experiments were conducted for three different reaction temperatures. The results are presented later on in this report. These results indicate favorable reduction of NO by reburning with methane, consistent with our computer projections, and hence, present a good base for investigating further on nitric oxide reburning with other fuels as well.

Experiments on nitric oxide reburning with i) a combination of methane and acetylene, and ii) a combination of methane and ammonia, will be considered in the next quarter. The computer simulation results submitted in the Second Quarterly Report indicate further reduction of NO with the addition of acetylene/ammonia to methane as reburn fuel. These results will be verified experimentally in the ensuing quarter.

Appended also, are two papers accepted for publication in the Proceedings of the Fourth HBCU/PS Fossil Energy Technology Transfer Symposium (to be) held in Greensboro, NC during April 2-3, 1996. At the time of writing of this report, Dr. Kumpaty had already presented these papers at the Symposium.

EXPERIMENTAL FACILITY

Seven gas cylinders (He pure, 20.1% O₂/balance He, CO₂ pure, 20.1% CH₄ / balance He, 2.19% C₂H₂/ balance He, 0.352% NO/ balance He, 0.36% NH₃/ balance He), fitted with respective pressure regulators were safely chained to the walls. The pressure regulators, valves and fittings for ammonia and nitric oxide had to be stainless steel due the corrosive nature of the gases. A quarter inch teflon tubing was employed for all connections. Using the proper Swage-lok fittings and ferrules, the gas cylinders were connected to the inlet of respective flow meters. The flow meters employed were of K-74 series purchased from King Instrument Company. The float valves for oxygen and NO flow were made of stainless steel, the valves for CO₂ and helium were made of sapphire and the others were made of carboloy. All these were chosen to suit the desired flow rates through the respective flow meters. Figure 1 shows the front view of the panel on which the seven flow meters are fitted.

Figure 2 illustrates the process of mixing of the gases from the seven cylinders before the gas mixture enters the reactor. First, all the gases, upon leaving the respective flow meters enter into the stainless steel manifold. However, in order to ensure homogeneous mixing, the gas mixture is drawn into a stainless steel chamber from the manifold. This chamber acts as the buffer vessel where proper mixing takes place. The gas mixture is then led to the reactor inlet through teflon tubing. Figure 3 shows the side view of the panel on which the flow meters and the buffer vessel is mounted.

Figure 1. Front view of the panel mounted with flow meters

Figure 2. Schematic of gas flow through flow meters, manifold and buffer vessel

Figure 3. Side view of the panel showing flow meters, manifold and buffer vessel

The teflon tubing of 0.25" dia is fitted onto the 1" OD ceramic reactor with adaptors on both ends of the reactor. Figure 4 shows the details of the inlet end adaptor. Notice the provision for future coal injection. Upon finishing the reburning experiments with homogeneous gas mixture, we will be testing various coals as reburn fuel.

Figure 5 shows a similar adaptor for the exhaust end of the ceramic reactor. Notice two ports on the adaptor- the 1/4" fitting was set for the gas flow and the 3/16" fitting for thermocouple insertion. The Omega high temperature probe measures the gas temperature in the reactor about 3" inside the furnace. The ceramic reactor which is 26" long, is enclosed by a 1200 C Thermolyne furnace (15.5") in its middle portion. Presently the reactor-furnace assembly is stationed horizontally for convenience. It will be held vertically for heterogeneous reactions with coal reburning. The temperature probe is connected to a HH12 digital thermometer.

The gas at the exhaust end of the reactor is led to the NOx analyzer via gas dryer. The dryer ensures removal of moisture. Teflon tubing of sufficient length was used to allow the cooling of gas mixture to the temperature range acceptable for the operation of the analyzer. This is to protect the NOx analyzer, which is the costliest piece of equipment. The analyzer needs oxygen supply for the ozonator which is accommodated from the same oxygen cylinder used in the reburning experiments by employing a T connection. The gas, upon analysis, leaves the analyzer and vents through the exhaust hood of the building.

Figure 4. Ceramic reactor inlet end adaptor with leak proof connections

Figure 5. Ceramic reactor exhaust end adaptor with the thermocouple

RESULTS AND DISCUSSION

As many as 15 runs were conducted on nitric oxide reburning with methane. First, the furnace was turned on and with its temperature set point control, a particular furnace temperature was set. Slowly, helium, oxygen and carbon dioxide cylinders were opened and the flow levels calculated for a particular reburning stoichiometric ratio (SR2) were set. Oxygen simultaneously flowed through NOx analyzer. The analyzer was turned on and allowed to warm up. The parameters on the analyzer were checked until they reached the normal operating conditions. Then, the nitric oxide flow was adjusted so as to read 1000 ppm on the NOx analyzer digital readout. The Omega probe measured the gas temperature inside the reactor which was digitized on the thermometer readout. When all the flow parameters were stable and the desired gas temperature was reached, methane was introduced according to the calculated flow rate for the particular SR2 in question. Instantly, the NOx output decreased and once it reached a steady value, the reading was recorded. A constant check on gas leaks was crucial to the success of the experiments.

When the furnace was set at 1130 C, the gas temperature 3" inside the reactor read 1092 C. The experiment was performed for five SR2 values, namely, 0.8, 0.85, 0.9, 0.95 and 1.0. The total flow rate of the gas mixture was 1950 cc/min. Given in Table 1 are the various flow rates of pure gases calculated for NO reburning with methane. Table 2 lists the actual flow rates, adjusted to accommodate varying gas proportions (that is, percent concentrations) in the cylinders such as oxygen/He, methane/He and NO/He. These were calibrated for rotameter scales and fed through the respective flow meters. The steady readings on the NOx analyzer before addition of methane and after addition of methane were recorded. The experimental results are shown in Table 3.

Table 1. Simulated flow rates of various pure gases for NO reburning with methane

SR2	CO ₂	O ₂	CH ₄	NO	He
0.80	315.9	36.3	69.9	1.95	1526.0
0.85	318.4	36.8	55.1	1.95	1538.0
0.90	320.7	37.0	41.7	1.95	1548.9
0.95	322.7	37.3	29.6	1.95	1558.7
1.00	324.6	37.5	18.5	1.95	1567.7

Table 2. Adjusted flow rates accounting for gas proportions (% concentrations) in the cylinders

SR2	CO ₂	O ₂	CH ₄	NO	He
0.80	315.9	180.4	347.6	553.4	1526.0
0.85	318.4	181.8	273.9	553.4	1538.0
0.90	320.7	183.1	207.4	553.4	1548.9
0.95	322.7	184.3	147.2	553.4	1558.7
1.00	324.6	185.4	92.2	553.4	1567.7

Later, the furnace temperature was adjusted to the maximum, that is, 1200 C, which in turn, maintained the gas mixture in the reactor at a temperature of 1153 C. Five runs were conducted at this set temperature for the flow rates mentioned above (five SR2 conditions). Similar procedure was repeated for another temperature setting with furnace temperature of 1050 C and a corresponding reaction temperature of 1010 C. All the results are summarized in Table 3.

Table 3. Experimental results on NO reburning with methane

Reburning SR2	Furnace T 1200 C Gas T 1153 C		Furnace T 1130 C Gas T 1092 C		Furnace T 1050 C Gas T 1010 C	
	NO in	NO out	NO in	NO out	NO in	NO out
0.8	998	5	1012	14	914	89
0.85	963	3.7	1018	10.3	922	72
0.9	960	2	1012	6	941	55
0.95	955	449	956	270	944	352
1.0	945	777	1018	852	1027	830

NO concentrations are measured in ppm.

It is apparent from Table 3 that the reburning stoichiometric ratio in the neighborhood of 0.9 is optimum for nitric oxide reduction and this trend is in line with the predictions reported earlier. Similarly, the reaction temperature of about 1100 C as reported in the numerical simulation results is verified to be an optimum for NO reduction. The reduction at 1092 C as well as 1153 C is significant compared to reduction at 1010 C. In fact, these experimental results indicate better reduction than the numerically predicted values; for e.g., a maximum reduction to 46 ppm from 1000 ppm was predicted through computer projection at SR2=0.9 for NO reburning with methane at 1100 C while the experiment shows nitric oxide reduction to 6 ppm from 1012 ppm at 1092 C for the same SR2. These results are very encouraging and support further investigation on reburning effectiveness of various fuels, in both homogeneous and heterogeneous reaction setting.

Extensive calculations were carried out on the reactor design during the third quarter, to show that the gas would attain a temperature of 1100 C inside the reactor. It was experimentally verified during NO reburning with methane, as reported in Table 3. The gas temperature inside the reactor, measured 3" inside the furnace (or approximately 5" from the center of the furnace length) is about 40-50 C lower than the set furnace temperature. Hence, the gas temperature could even be less than 20 C lower at the middle portion of the furnace/reactor compared to the set furnace temperature. The measurement of gas temperature served as an important tool in establishing the proper reaction conditions in nitric oxide reburning.

PROJECTIONS

In the ensuing quarter, acetylene will also be introduced along with methane as reburning fuel and NO_x reduction will be investigated for various SR2 conditions. Similar experiments will be conducted for ammonia as reburn fuel addition to methane. An effort will be made to procure coal samples for future heterogeneous reactions. Also, the design of an effective coal feeder will be another item of our perusal.

RECENT PUBLICATIONS

Enclosed in the following pages are two papers presented at and published in the proceedings of the Fourth HBCU/PS Fossil Energy Technology Transfer Symposium held in Greensboro, NC during April 2-3, 1996.