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

Technical Progress Report
Seventh Quarter
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INTRODUCTION

During this quarter (July-August 1996), the experiments for nitric oxide reburning with a combination of methane and ammonia were conducted successfully. This marked the completion of gaseous phase experiments. Preparations are underway for the reburning studies with coal. A coal feeder was designed to suit our reactor facility which is being built by MK Fabrication. The coal feeder should be operational in the coming quarter. Presented here are the experimental results of NO reburning with methane/ammonia. The results are consistent with the computational work submitted in previous reports.

REBURNING EXPERIMENTS WITH METHANE/AMMONIA

EXPERIMENTAL

The experimental procedure employed for nitric oxide reburning with methane/ammonia was very similar to the one employed for reburning with methane/acetylene combination. First, the furnace was started and with its temperature set point control, a furnace temperature in the neighborhood of 1140 C 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 the 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 to 1000 ppm which was expressed on the NOx analyzer digital readout. The Omega probe measured the gas temperature inside the reactor which was digitized on the thermometer readout. The furnace temperature was adjusted slightly so as to maintain the reactor gas temperature at 1100 C. When all the flow parameters were stable, methane and ammonia were 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. The procedure was repeated for various SR2 values (0.8-1.0) as well as for two reburn fuel combinations of methane and ammonia (98/2 and 96/4). The model results steered the choice of input conditions in this experimental study on the reburning effectiveness of methane/ammonia.

While the gas temperature 3" inside the reactor was steady at 1100 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 98/2 fuel combination of methane and ammonia. 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, ammonia/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 reburn fuel and after addition of reburn fuel were recorded. The experimental results for 98/2 combination of methane/ammonia, with introduction of methane only, methane and ammonia together and ammonia only (by closing methane feed and increasing ammonia to the allowable maximum through the rotameters) are shown in Table 3. The flow rates of pure gases and the adjusted rates were calculated for the 96/4 combination of methane and ammonia and the entire cycle of experiments was performed at SR2 values of 0.85, 0.9, 0.95 and 1.0. The results of these experiments are presented in Table 4.

Table 1. Simulated flow rates of various pure gases for NO reburning with 98/2 combination of methane/ammonia

SR2	CO_2	O_2	CH_4	NH_3	NO	He
0.80	315.8	36.2	69.3	1.41	1.95	1525.3
0.85	318.3	36.5	54.6	1.11	1.95	1537.5
0.90	320.6	36.8	41.4	0.84	1.95	1548.5
0.95	322.6	37.0	29.3	0.60	1.95	1558.4
1.00	324.5	37.4	18.4	0.38	1.95	1567.5

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

SR2	CO_2	O_2	CH_4	NH_3	NO	He
0.80	315.8	180.4	344.9	151.4	553.4	404.2
0.85	318.3	181.8	271.8	306.2	553.4	318.5
0.90	320.6	183.1	205.8	445.9	553.4	241.2
0.95	322.6	184.3	146.0	572.5	553.4	171.1
1.00	324.5	185.3	91.5	687.9	553.4	107.3

Table 3. Experimental results on NO reburning with reburn fuel of 98% methane and 2% ammonia

Gas Temperature 1100 C

SR2	NO in	NO_{out}^{*} methane only	NO_{out} CH_4 and NH_3	NO_{out}^{*} ammonia only
0.8	1020	43	**24**	885
0.85	1010	37	**24**	860
0.9	990	31	**22**	845
0.95	1022	310	**275**	934
1.0	1080	940	**905**	1045

NO in/out Concentrations are measured in ppm.

*Not representative of SR2 value. Based on cutting off one or the other reburn fuel from the reaction mixture.

Table 4. Experimental results on NO reburning with reburn fuel of 96% methane and 4% ammonia

Gas Temperature 1100 C

SR2	NO_{in}	NO_{out}* methane only	NO_{out} CH_4 and NH_3	NO_{out}* ammonia only
0.85	994	38	**34**	697
0.9	1012	32	**22**	789
0.95	1000	431	**352**	861
1.0	1014	928	**865**	929

NO in/out Concentrations are measured in ppm.

*Not representative of SR2 value. Based on cutting off one or the other reburn fuel from the reaction mixture.

DISCUSSION

It can be seen from Table 3 as well as Table 4 that for the case of methane only as reburn fuel, NO reduction increases with increase in SR2 ratio until the optimum SR2 value of 0.9. The reduction is not as high for the cases of SR2 > 0.9. This behavior was documented in the previous report and is consistent with the numerical predictions carried out earlier in the program.

For the case of 98% methane and 2% ammonia (Table 3), a significant NOx reduction is observed. The inlet concentration of NO (1000 ppm) reduces to lower twenties in the ppm level for the SR2 values upto 0.9. The reduction is 73% at SR2=0.95 and only 16.2% at SR2=1.0. These experimental results of NO reduction with a combination of methane and ammonia follow the same trend predicted computationally.

It can be noticed from Table 4 that that NO reduction is similar (to the above trend) for 96/4 combination of methane/ammonia. The maximum reduction occurs at SR2=0.9. The reduction is less at higher SR2 ratios: 64.8% at 0.95 and only 14.7% at 1.0. However, comparing the levels with the introduction of methane only, it can be inferred that a slight addition of ammonia favors the NOx reduction further by strengthening the reductive effectiveness of methane. It can be further observed from Tables 3 and 4 that the additional effect of ammonia on NOx reduction is more pronounced at SR2 > 0.9 than SR2 < 0.9. This is due to the fact that the methane-NOx reaction is not close to the equilibrium in the former case (SR2 > 0.9) than the latter case.

Also shown in Tables 3 and 4 is the exit concentration of NO when methane feed was cut off and only ammonia was used as the reburn fuel. This was deliberately planned to see the performance of ammonia as a primary reburn fuel. The reduction of nitric oxide was not much, a maximum of 14.6% for 98/2 run and about 22% for 96/4 run. Thus it was concluded that the use of ammonia in small quantities is helpful in NOx reduction chiefly as a reburn fuel additive to methane.

The above findings are significant in terms of the industry needs. With methane as a reburn fuel, the narrow operating window calls for precise cascade control between the primary zone combustion feed inlet, the reburning zone methane inlet and the NOx analyzer in order that the NOx emissions be within permissible limits. However, with the addition of acetylene or ammonia to methane as reburn fuel, the NOx emissions will be within permissible limits as long as a set point control is given to the methane/acetylene or methane/ammonia reburning feed inlet not to exceed the SR2 of 0.9. With the latter case, the operation is easier to keep the NOx emissions within limits even if there arise some changes in the primary zone combustion feed inlet.

COAL FEEDER AND FUTURE WORK

The coal feeder was designed with the following specifications: coal tube, 12mm internal dia glass; outer tube, 1" SS tubing; piston, SS with O ring seal; piston rod, 1/8' SS; end seals, swagelok type SS fittings with teflon ferrules; feed screw, ½" dia X 20 TPI; drive connection, acme nut type; linear bearings, Grainger 2X567; shaft bearings, permanently lubricated 2X897; linear shafts, ½" polished rod; frame plates, ½" aluminum; travel stops, SS ring 1L636; drive motor, Dayton 4Z536; flexible motor coupling; outlet tube, 1/8' SS tube. The design was provided to MK Fabrication and the coal feeder is being built. At the facility, the furnace-reactor setup is being adjusted to accommodate the coal supply into the reactor by the use of the coal feeder. The details of design and operation of coal feeder will be discussed in the coming quarterly report. It is expected that the reburning experiments with coal will be initiated in the next few months.