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

Technical Progress Report
Eleventh Quarter
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INTRODUCTION

Several experiments were conducted during this quarter to study the NO_X reduction effectiveness of lignite coal, activated carbon and catalytic sites such as calcium sulfide and calcium carbide. While some of the coals/chemicals could be fed easily, some needed the mixing with silica gel to result in a uniform flow through the feeder. Several trial runs were performed to ensure proper feeding of the material before conducting the actual experiment to record NO_X reduction. The experimental approach has been the same as presented in the past two quarterly reports with the coal reburning experiments. Initial reduction is achieved through methane addition for SR2=0.95 conditions and then coal or the catalyst is introduced to see if there is further reduction. Presented below are the results of the experiments conducted during this quarter.

REBURNING WITH MONTANA LIGNITE

The difficulty of feeding the DECS-25 coal sample (Montana lignite coal, Richland county), reported in the previous quarter, was solved by mixing the sample with silica gel, on a 50-50 weight basis. The coal bed did not get packed and the uniform feeding was achieved in this process. After ensuring the free flow of the coal-silica gel mixture, the coal feeder assembly was incorporated in the experiment and the experimental conditions for methane reburning were set. The nitric oxide level was set at 1000 ppm as in all the previous experiments. While methane was introduced (as per SR2=0.95 conditions), the NO_X reduced to 461 ppm. A continuous feeding of the coal-silica gel mixture was verified and further reduction was checked. The reduction was observed for two feed rates, referred to hereafter as Rate I (speed 5 on the variable speed motor of the coal feeding mechanism) and Rate II (speed 7 on the variable speed motor). The feed rates presented below the table represent only the amount of coal fed per minute while the same amount of silica gel is also fed in that time. So the feed rate of the mixture is twice the feed rate of the coal. Table 1 lists NO_X readings on the analyzer read-out.

Table 1. NOX reading during reburning with methane and coal (DECS-25 sample), ppm

Initial NOX set at 1000 ppm.

SR2	With methane only	With methane and coal
0.95	461	39* 16+

*Rate I: 0.114 gm/min of DECS-25 coal
+Rate II: 0.13 gm/min of DECS-25 coal

It is apparent that the introduction of coal aids the reduction of NO_X by reburning with methane at SR2=0.95 quite considerably. The level of reduction achieved by methane alone at SR2=0.9 (up to <50 ppm out of 1000 ppm) can now be obtained at SR2=0.95 with the introduction of coal, as a result of coal char gasification, thereby enhancing NO_X reduction.

REBURNING WITH ACTIVATED CARBON

Having seen favorable NOx reduction with several coal samples, the research team felt the need to attempt reburning with activated carbon, as an appropriate closing point for coal reburning experiments. Since this is a pure carbon with less than 1% impurities, the best possible reduction was expected. The results were in line with the expectation. The activated carbon was the most freely flowing and hence, the easiest to feed, of all the samples tried. Presented in Table 2 are the NO_X readings on the analyzer read-out.

Table 2.NO_X reading during reburning with methane and activated carbon, ppm

Initial NO_X set at 1000 ppm.

SR2	With methane only	With methane & activated carbon
0.95	447	7* 7+

*Rate I: 0.154 gm/min
+Rate II: 0.232 gm/min

As seen in the case of lignite coal, activated carbon reburning experiments confirmed significant NO_X reduction by char gasification. It can be concluded from the results presented above that the reduction achieved by activated carbon (upto 7 ppm) in aiding reburning with methane is very significant as a NO_X reduction strategy in industrial applications. Increasing the amount of activated carbon (as shown by Rate II) did not matter once the reduction was achieved. It can be safely said that the presence of activated carbon boosts heterogeneous reactions with the gas mixture inside the reactor maintained at 1100 C. Modeling surface reactions in this setting would be a useful study for future.

SURFACE CATALYZED REBURNING

An important phase under this grant instrument is to conduct surface catalyzed reburning studies. In deliberation with the concerned project personnel at the Department of Energy, it was decided that calcium sulfide and calcium carbide would be used to conduct reburning studies and estimate their catalytic effect on NO_X reduction under pulverized coal combustion conditions.

a) Study with CaS

Calcium Sulfide, as obtained from Aldrich, is in powdered form and tends to pack very quickly. Obviously, it could not be fed properly into the reactor. A procedure similar to that used for the Montana lignite coal was employed. An equal amounts of CaS and silica gel were mixed together uniformly and the mixture was placed in the plexi-glass chamber of the feeder. The feeder and the feeding mechanism are the same as those used for feeding coal. Upon setting all the experimental conditions for reburning with methane at SR2=0.95 and achieving NO_X reduction from 1000 ppm to 453 ppm, the uniform feeding of the CaS- silica gel was maintained. Speed 9 on the variable speed motor was employed to allow for the maximum feed rate of 0.12 gm/min of CaS (or, 0.24 gm/min of CaS- silica gel mixture). With the calcium sulfide supply at the rate of 0.12 gm/min, NO_X reduced to 205 ppm, increasing the reduction from 55% to 80% overall. This reduction proves that CaS offers catalytic surface, thereby enhancing an additional 25% NO_X reduction. The observations are presented in Table 3.

Table 3. NO_X reading during reburning with methane and calcium sulfide, ppm

Initial NO_X: 1000 ppm. Reaction temperature: 1100 C.

SR2	With methane only	With methane & calcium sulfide
0.95	453	205[@]

[@] Feed Rate of CaS: 0.12 gm/min

b) Study with CaC_2

Calcium carbide, unlike calcium sulfide, was received as 8 mm thick pieces and had to be ground to fine particles in order to be fed through the coal feeder. There was no problem of the bed being packed and the flow into the reactor was uniform. Calcium carbide was also purchased from Aldrich. Presented in Table 4 are the NO_X readings on the analyzer readout.

Table 4. NO_X reading during reburning with methane and calcium carbide, ppm

Initial NO_X: 1000 ppm. Reaction Temperature: 1100 C.

SR2	With methane only	With methane & calcium carbide
0.95	467	152[*] 113[+]

[*]Rate I: 0.106 gm/min
[+]Rate II: 0.122 gm/min

The feed rate of 0.106 gm/min (Rate I) for calcium carbide shows a significant NO_X reduction, namely, 53% with methane alone and 85% with both methane and calcium carbide. The feed rate of 0.122 gm/min (Rate II) shows further reduction, up to 89%. This experiment further confirms our previous conclusion that calcium associated compound offers a catalytic surface thereby abetting NO_X reduction significantly. Since Rate II in this experiment is comparable to the feed rate of calcium sulfide that yielded reduction up to about 80%, it can be further concluded that calcium carbide seems to show higher catalytic effect on NOx reduction than calcium sulfide.

PROJECTIONS

With the end of this quarter (11th), the experimental activities planned under this grant are successfully completed and the remainder of the grant period will be used to pool all the information together and prepare the final report. The final report will document all the numerical modeling and the experimental studies conducted under this grant instrument since its initiation (02/01/95). There will not be a separate report for the last quarter.