MINIMIZATION OF NO EMISSIONS FROM MULTI-BURNER COAL-FIRED BOILERS

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Program Overview

The focus of this program is to provide insight into the formation and minimization of NOx in multi-burner arrays, such as those that would be found in a typical utility boiler. Most detailed studies are performed in single-burner test facilities, and may not capture significant burner-to-burner interactions that could influence NOx emissions.

Our approach is to investigate such interactions by a combination of single and multiple burner experiments in a pilot-scale coal-fired test facility at the University of Utah, and by the use of computational combustion simulations to provide insight into the experimental results and to evaluate full-scale utility boilers. In addition, fundamental studies on nitrogen release from coal will be performed in support of the modeling effort. Improved submodels describing transformations of both volatile nitrogen species and char nitrogen species will be developed.

The program is broken into four main tasks, and reporting will be divided into these main areas:

1- Fundamental studies on nitrogen release from coal. These studies will be used to enhance the predictive capabilities of the combustion simulations. Studies focusing on secondary coal pyrolysis will be carried out at Brigham Young University, and studies focusing on char nitrogen will be performed at the University of Utah.

2- Comprehensive modeling of burner arrays. This task will be performed by Reaction Engineering International and the Unversity of Utah.

3- Pilot-scale optimization of multi-burner arrays. This task will be carried out by the University of Utah.

4- Technology transfer. This task involves coordination with utility consultants who will provide oversight of the research program.

FUNDAMENTAL STUDIES ON NITROGEN RELEASE FROM COAL

Nitrogen Transformations during Secondary Coal Pyrolysis

Introduction

Reduction of NO_x emission is an important environmental issue in pulverized coal combustion. Final emissions of NO_x are strongly affected by the nitrogen release during devolatilization, which is the first stage of coal combustion. The most cost-effective approach to NO_x reduction is air-staging which can also operate with additional down-stream techniques such as reburning [1]. Air staging promotes the conversion of NO_x precursors (HCN, NH_3, etc.) to N_2 by delaying the oxygen supply to the greatest extent when those nitrogen species are released during devolatilization. Such a delay gives the primary volatiles a chance to undergo secondary reactions, including tar cracking and soot formation. Secondary reactions of volatiles largely determine the fate of the ultimate NO_x production from pyrolysis, therefore a detailed investigation into the transformation of nitrogen species during secondary reactions and effects of soot on nitrogen release is critical for design and implementation of new pollution control strategies. Current nitrogen models (including the CPD model at BYU) only simulate the nitrogen release during primary pyrolysis, which happens at low temperatures. This project helps to build a nitrogen release model that accounts for secondary reactions and the effects of soot at temperatures relevant to industrial burners.

Objectives and Approach

The objectives of this project are: (1) to effectively determine the mass release and soot/tar yield of four selected coals at different temperatures and residence times; (2) to quantify the distribution of nitrogen species among char, tar/soot and gas phase during secondary reactions; (3) to investigate the effects of such factors as coal rank, temperature, residence time on nitrogen release during secondary pyrolysis; (4) to produce a secondary nitrogen release model based on the current CPD model.

Accomplishments

Accomplishments for the past reporting period include the following:

1. Completion of a set of experiments in the Flat Flame Burner (FFB) that includes 4 coals, 4 temperatures and 4 residence times. These experiments provided char and tar/soot samples for elemental analysis, ICP analysis and diffuse reflectance analysis on FTIR.

2. Demonstration of low temperature (as low as 1050 K) experiments in the FFB.

3. Completion of several test runs in the drop tube reactor (HPCP) that included 2 coals. Some minor changes to the HPCP were made and temperature profiles were measured for two proposed conditions.

4. Accurate quantification of HCN, NH_3, light hydrocarbons and other significant N species in gas phase was performed using FTIR spectroscopy.

5. N distributions among char, tar/soot and gas phase was determined at 4 temperature and 4 residence time conditions.

FTIR Analysis of N Species in Gas Phase

Measurement of gas phase N species in the FFB has been a big challenge in this project. In the FFB, the large quantities of hot burning gas provide the necessary high temperature and high heating rate used to study coal N release. In order to attain single particle reactions, the coal loading is also kept very low (about 1g/hour). The actual concentrations of N species fall in the range of parts per billions in the pyrolysis experiments.

Quantification of gaseous N species (only HCN and NH_3) during pyrolysis was previously attempted using an HCN monitor. However, it was recently shown that the HCN monitor could not be used to accurately quantify HCN because of a huge drift of the data, resulting in standard deviations as high as 500%. NH_3 measurement was also shown to be unreliable using the HCN monitor. A high resolution Gas Chromatography had also been tested, however, the detection limit of the GC is only up to high ppm-levels.

Application of FTIR on trace gas analysis in combustion system have been reported by several researchers [2][3][4][5]. The techniques in IR spectroscopy can be used for most common combustion gas measurements, and at the same time eliminate the complexity and reliability problems experienced with systems employing multiple individual gas analyzers, each with their own detectors. However, since the reported trace gas concentrations are usually in the range of ppm (from 5 ppm to several hundred of ppm), accurate measurement of trace gas in the low ppb range is much more difficult.

A BOMEM MB155 FTIR coupled with a 9.75m multi-reflection gas cell was successfully used to perform on-line measurements of ppb-level HCN, NH_3, hydrocarbons and other significant species in the gas phase. The schematic of the sampling system is shown in Figure 1. Spectra were collected with a resolution of 1.00 cm^{-1} and spectral range of 500 cm^{-1}–4000 cm^{-1}. The pyrolysis gas from the sampling line was passed into the gas cell after passing glass filters. IR scans were made after the gas cell was purged for about 5 minutes. By using a liquid N_2-cooled MCT detector and 1-wavenumber resolution, the detection limit of the FTIR can be as low as 50 ppb for some gases (including NH_3, C_2H_4 and C_2H_2). FTIR has been proved to be the most effective way to measure the ppb-level gas species in this project.

A CO flame (with a small amount of H_2) was used to perform the experiments, in order to minimize the amount of steam in the post-flame gases. Even with the large reduction in steam concentration, it is quite difficult to obtain IR measurements in a harsh environment containing 15% CO_2, 25% CO and small amount of water. All of these species are extremely strong IR absorbers, which can greatly interfere with the measurement of other weakly absorbing species. The data collected are also reliable and reproducible. Figure 2 shows sample spectra of HCN and NH_3 from coal pyrolysis in the CO flame. Figure 3 shows the repeatability of some gas measurements. All measured gas concentrations are stable except NH_3. The drift of NH_3 measurement is due to the so called "memory effect" which is caused by selective adsorption and desorption of NH_3 from the cell wall [6]. In real experiments, NH_3 was measured immediately after the gas cell was filled in about 40 seconds (10 scans) in order to minimize the memory effect. Other gases were measured after 144 scans in order to get a better signal to noise ratio.

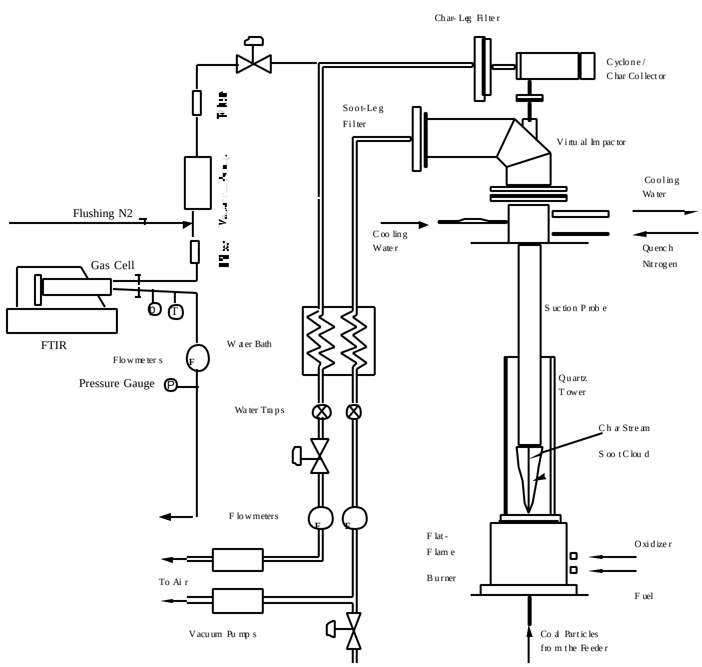

Figure 1. Experimental Apparatus.

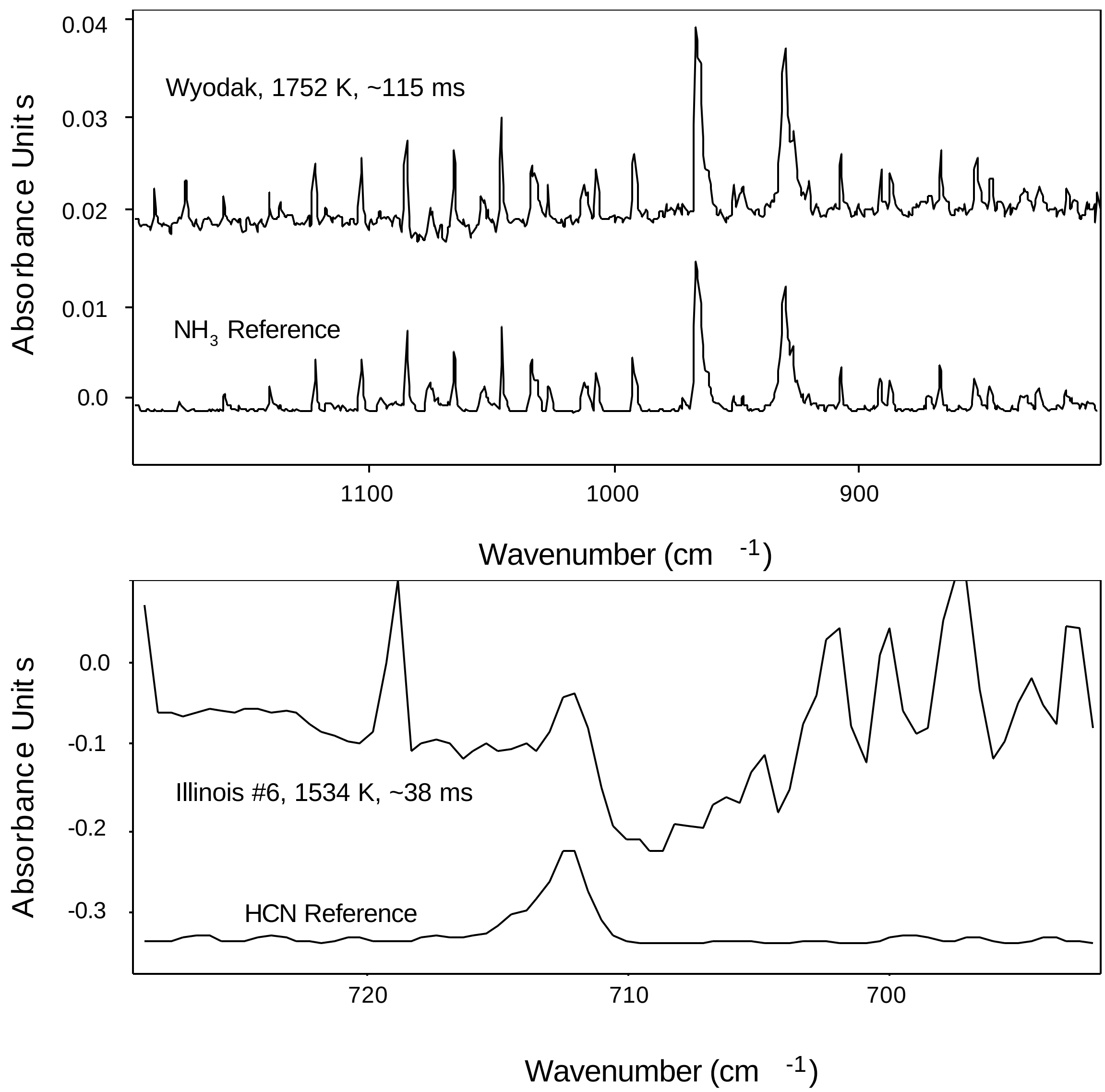

Figure 2. Comparison of measured and reference spectra for HCN and NH_3.

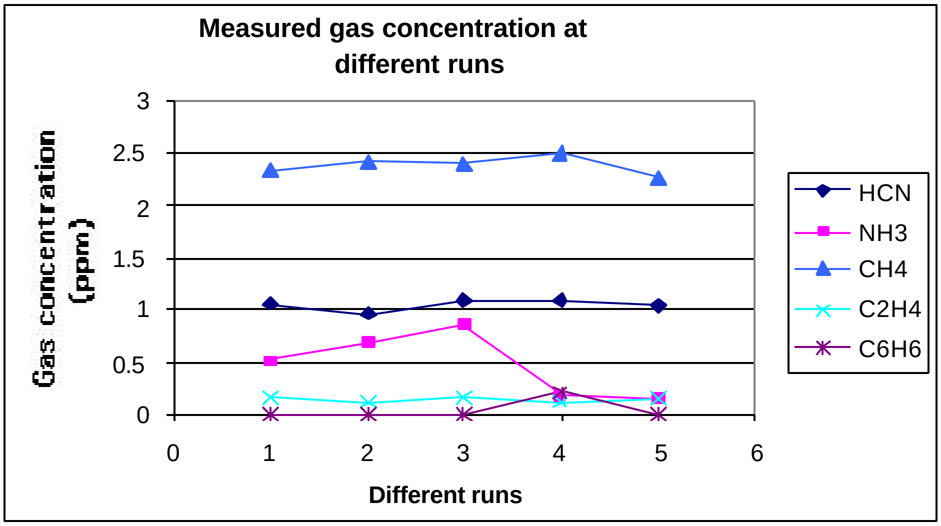

Figure 3. Duplicate FTIR measurements of major pyrolysis gases

Low-temperature FFB Experiments

The FFB has been used extensively at BYU for several years in high temperature, high heating-rate pyrolysis and combustion experiments. The regular fuel used in the FFB is methane. Last year, the fuel was changed to CO because large quantities of water produced during CH_4 combustion will interfere with FTIR analysis. The flammability limit of methane is very narrow (from 5% to 15%, volume in air), so theoretical calculations shows that the minimum adiabatic flame temperature of methane flame is about 1650 K. Therefore, it was assumed that the FFB could only be used in high temperature experiments (> 1500 K). It was also assumed that low temperature experiments had to be conducted in the drop-tube reactor. However, it was recently found that there is another advantage to using a CO flame. The flammability limit of CO flame is much broader than a CH_4 flame (12.5% -74.2%, volume in air), which means that the flame can be sustained even at very fuel-rich conditions [7]. The calculated adiabatic flame temperature of CO is about 1000 K. Recent experiments have shown that it is easy to reduce the FFB flame temperature down to at least 1100 K, while sustaining a stable flat flame. Because the FFB is much easier to operate and less prone to break down than the HPCP, using the FFB to conduct low temperature experiments can save considerable time and cost. Since all the experiments are conducted in the same reactor, the pyrolysis data are also easy to compare. Three new temperature conditions were demonstrated and used in experiments to study the transition from tar to soot, which is characteristic of secondary pyrolysis reactions, as shown in Table 1.

Table 1
Temperature conditions used in FFB experiments

Condition:	1 low	2 low	3 low	4	5	6	7

	temperature	temperature	temperature				
Peak flame temperature	1159 K	1281 K	1411 K	1534 K	1618 K	1752 K	1858 K

Results of FFB Experiments

Secondary nitrogen release data on the nitrogen distribution among char, tar/soot and various gas species are shown in Figures 4 and 5. Char and tar/soot were collected at different temperatures, with residence times ranging from 14 ms to 45 ms. The pyrolysis gas nitrogen species concentrations were quantified by the above-mentioned FTIR analysis. From the figures, it can be found that the nitrogen balance is excellent for most conditions, usually within 10% deviation. At higher temperatures, the high yields of NH_3 seem suspicious for some low rank coals (Figure 6). However, it has to be pointed out that NH_3 yield presented here were based on the highest NH_3 concentrations recorded in those conditions. These preliminary measurements were made before the memory effects were identified and more reliable procedures were adopted

Future Plans

Future plans include completing a series of pyrolysis experiments on selected coals in the FFB and HPCP. Analysis of data from these experiments will permit the examination of nitrogen release behavior during the second stage of coal pyrolysis. The effects of temperature, residence time, coal rank on secondary nitrogen release will be investigated to provide helpful information for future pollution control design. The effect of soot on nitrogen transformation is also an indispensable part in this project. A secondary nitrogen release model will be proposed based on current experimental data and other important data from the literature. This model will be incorporated into the CPD model [8], which will enable simulation of the volatile release, nitrogen release, and secondary pyrolysis that occurs in combustion processes.

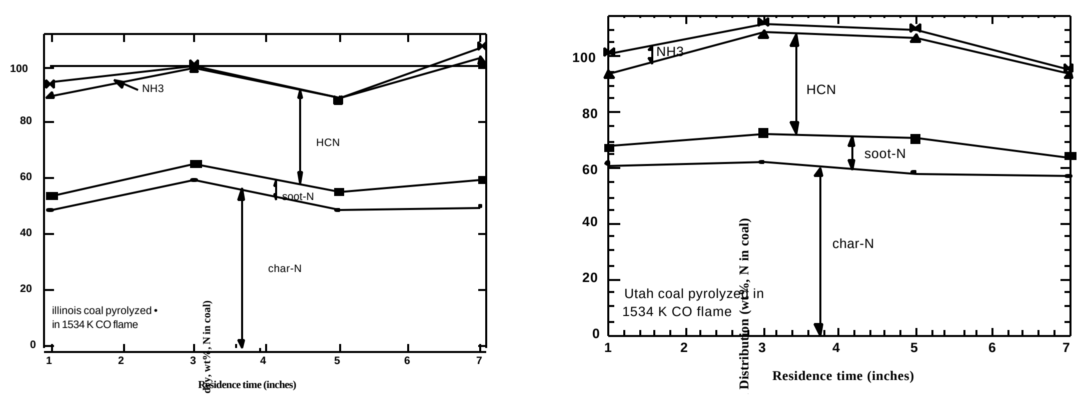

Figure 4. Cumulative nitrogen distribution for the Illinois #6 and Utah coal pyrolysis experiments in the 1534 K CO flame.

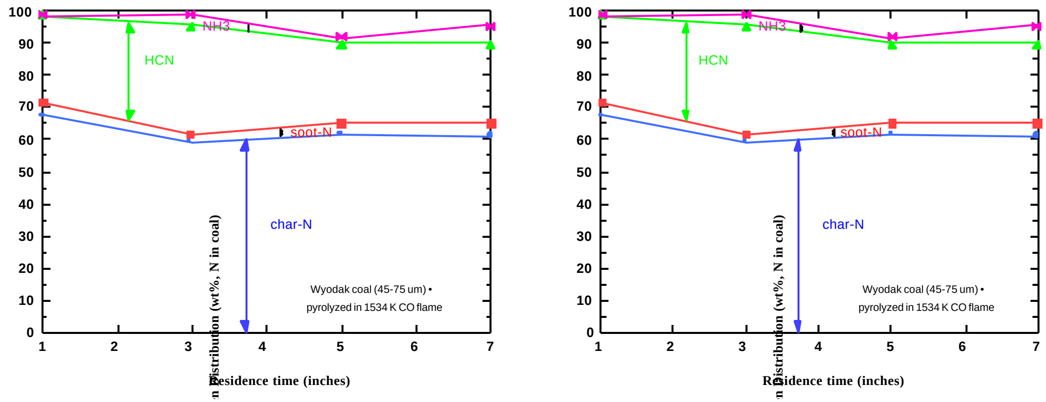

Figure 5. Cumulative nitrogen distribution for the Wyodak subbituminous coal and KR lignite pyrolysis experiments in the 1534 K CO flame.

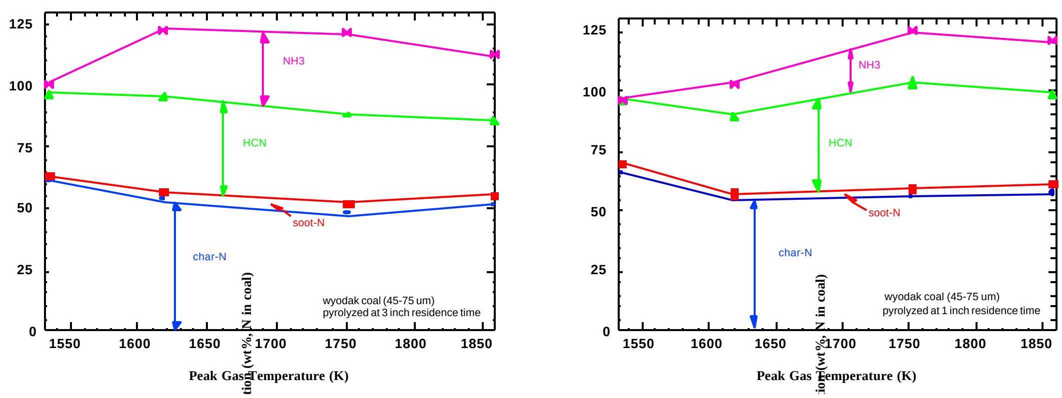

Figure 6. Temperature effects on cumulative nitrogen distribution for the Wyodak subbituminous coal and KR lignite.

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Pilot-Scale Studies: Preparations for Multiburner Firing

As the burners were being fabricated, the design for the installation and operation of the multiple burners was started. A vertical arrangement of three burners was decided to be the desired configuration for the burners (Figure 1). In this arrangement, there will be both mixing from burner to burner and buoyancy effects. The burners centerlines were spaced on the same distance as the L1500 sample ports. This will provide the best opportunity to obtain centerline emissions profile along the length of the furnace to determine the mixing characteristics. The burner quarl was designed with a 35^{o} angle and L/D=1 to simulate a full-scale boiler. Each burner will have 2 one-inch holes angled from the outside of the shell to the quarl for peepers and ignition sparkers. An additional peeper will be placed down the barrel of each burner for gas flames during furnace heat-up and overnight conditions.

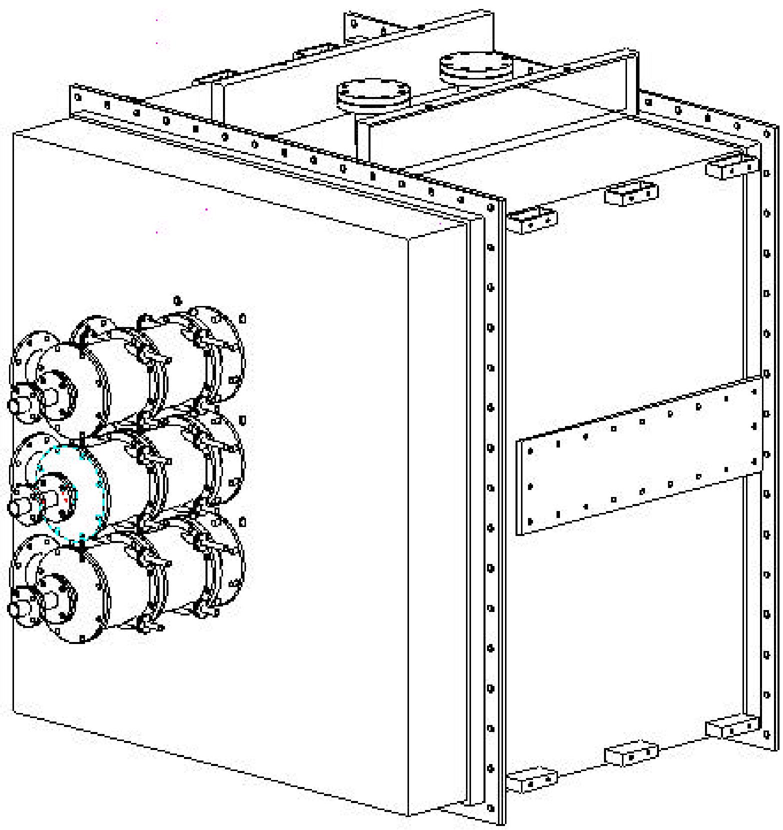
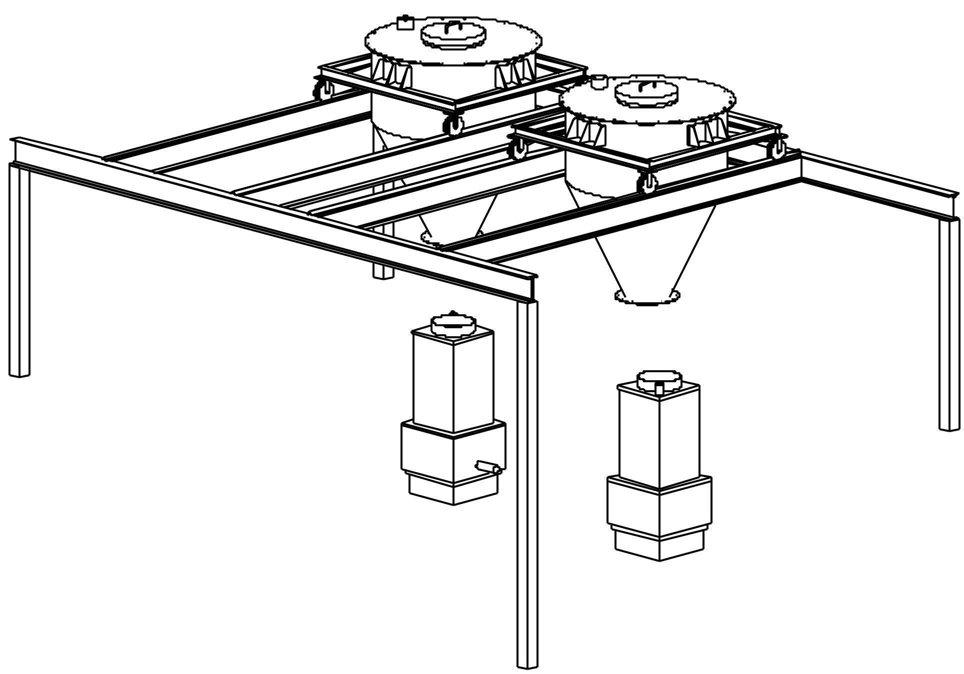


Figure 1. Burner Arrangement with Section 1 of the L1500.

Each burner has primary air, secondary air, tertiary air, natural gas, and coal streams which are measured and controlled independently from the other burners. The secondary and tertiary air inputs are independently controlled for both flow and preheat temperature. The flows are measured by V-Cones and are controlled by control valves. The temperatures are controlled with electric heaters and SCR's. The natural gas streams are controlled by mass flow controllers, which were sized to control the flow for low stabilization gas flows as well as heat-up and overnight conditions. Primary air streams are measured with V-Cones and controlled by control valves before going through an eductor to carry the pulverized coal to the burner. The coal is fed into the eductors with screws from loss-in-weight feeders.

The L1500 furnace previously had only one coal feeder and associated hoppers. In order to accurately control additional coal streams, two additional loss-in-weight feeders were purchased. In addition, new hoppers and slide valves were designed and purchased. The location of the new feed system made for some interesting design problems. The pulverized coal is loaded into the hoppers from bulk solids bags using the building crane. However, the crane stops short of the hoppers, and therefore a trolley was designed to bring the hoppers to the crane (Figure 2). When a hopper is empty, the hopper will be disconnected from the feeder, through a soft connection, and rolled out to where the crane can fill the hopper from a



new coal bag.

Figure 2. New coal feed system with moveable hoppers and feeders.

Currently, the burners are fabricated and assembled. The burner plate is fabricated and refractory will be poured next week. The coal feed system structure is fabricated and ready for the hoppers, which are in fabrication. The natural gas mass flow controllers and the loss-in-weight feeders have been ordered and we are waiting for them to arrive. All of the secondary and tertiary air flow measurement and control, along with the heaters, are in place. Primary air flow measurement and control equipment have arrived and will be installed in the next couple of weeks. All wiring, plumbing and coding will be performed as the equipment is installed.