Char Burnout Kinetics in an Entrained Flow Reactor

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Understanding char burnout kinetics in coal combustion is critical for utility emissions control and subsequent fly ash usage in the cement industry. Using a commercial CFD code (Fluent), a numerical model of coal burning in the UC Irvine entrained flow reactor (EFR) was created to evaluate several common char burnout kinetic modeling approaches. The CFD simulations included appropriate convective and radiative heat transfer, fluid dynamics, as well as coal/ash particle tracking and combustion chemistry. EFR experimental data was obtained for operating conditions adjusted to reproduce particle Lagrangian temperature and oxygen concentration time histories typically found in coal-fired utility boilers. The temperature profiles are measured at different axial locations in the EFR and agree well with the simulations. New measurement capabilities are being implemented to similarly confirm oxygen distribution in the EFR and to provide more detailed oxygen distribution inputs to the model. A particulate sampling probe is used to extract ash samples at different heights in the reactor to measure the evolution of loss on ignition (LOI). Measured LOI values are used to validate the model against predicted values. Reaction kinetics rates in the model are adjusted to bring agreement between calculated LOI and the measured values from the experimental results. The long-term objective of the work is to evaluate the performance of the EFR-validated kinetic model in predicting char burnout in a full-scale commercial boiler. In addition, a sensitivity analysis of one of the kinetics approaches, the Char Burnout Kinetics (CBK) model is presented.

Keywords: *Char Burnout Kinetics, Loss on Ignition, Entrained Flow Reactor.*

Introduction :

A major waste product in the combustion of coal and biomass is large quantities of ash. The amount of carbon in the ash reflects boiler efficiency and determines whether the fly ash is a sellable product. High amounts of unburned carbon in the ash from many power plants prevents its sale as a raw material to the cement industry. Consequently, optimizing utility boilers to produce low carbon in ash that can be recycled instead of landfilled represents an important environmental benefit. To predict the relationships between boiler operating conditions and residual carbon-in-ash, there is a need for improvements in representing char reactivity. Developing these improvements and providing more accurate predictions of residual carbon-in-ash is the focus of this research.

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One problem in coal combustion characterization is the great diversity of coals, as they originate from various raw materials and conditions of formation. Chemical analysis shows that the increasing carbon content and decreasing oxygen content of higher rank coal leads to higher calorific value. On the other hand, increasing rank is generally associated with lower reactivity and slower char burn off. Char burn out is the slowest stage in coal oxidation, where reactivity loss and extinction phenomena can take place. Decreases in reactivity reduce burning rates, even at high temperatures, in the later stages of char burnout. Prior studies have shown that these decreases in reactivity will be followed by extinction phenomena if mass and heat transfer conditions are favorable [1].

To simulate coal combustion processes, rate expressions are required that accurately determine the kinetics of the heterogeneous char oxidation reactions throughout the fuel's burning lifetime. Because there is little heat release associated with the final stages of char burnout, the kinetic rates during this period are not as heavily considered. For carbon-in-ash prediction, however, this slow oxidation phase is critical. Hence, the goal of this project is to obtain kinetic data for use in computer simulations to predict burning performance of coals in existing boilers and to assess the accuracy of the existing char burnout models, particularly in regards to predicting residual carbon in the ash. A representative environment is reproduced in an entrained flow reactor (EFR) by matching the available oxygen and temperature environment with that of a realistic full-scale boiler [2,3].

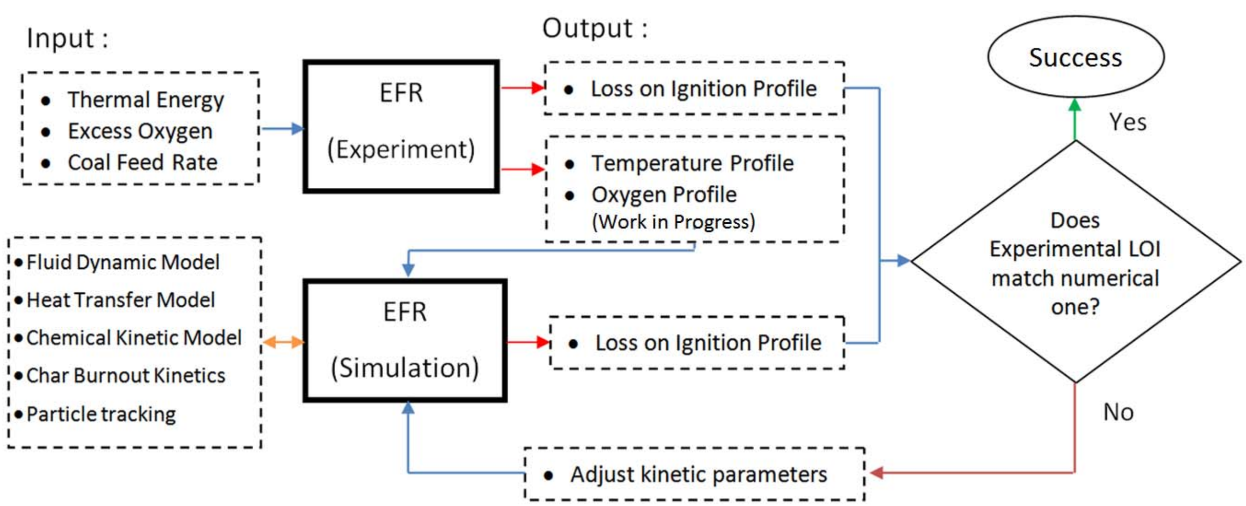

Figure 1 - Project approach flow chart

Figure 1 gives an outline of the project approach. After the EFR was designed and built [3], temperature profiles were measured and ash particles were collected to measure loss on ignition (LOI) as a function of residence time. Measured temperature and oxygen profiles inside the EFR are critical to ensure the accuracy of the model. The EFR model includes fluid dynamics, heat transfer, mass transfer, as well as coal/ash particle tracking and combustion chemistry. After the EFR model is validated, the next step is to select particle histories of interest and to compare measured LOI to LOI predicted by the model. This might enable the char burnout performance of a specific coal to be predicted from relatively simple, fast and inexpensive tests. Ultimately, the goal is to predict the performance using CFD.

There are other methods that have been used for studying coal combustion besides an EFR. For example, thermo-gravimetric analyzers (TGA) are fairly common. The main advantage of an EFR compared to TGA equipment is its heating rate. The rate of heating of particles in an EFR is estimated to be 10^4 to 10^5 K/s, while the heating rate of a TGA is generally 4 to 5 orders of magnitude smaller (15-25 K/min). Also, since the particle and environment temperature are not easily known in the TGA, valid kinetic calculations are difficult. Nevertheless, TGA results have been shown to be valid for comparing different types of coal [4].

In the EFR simulation, the simple FLUENT built-in kinetic-diffusion limited model and Fluent's internal CBK model, following the work of Hurt [1], are used for char burnout calculation. The simple kinetic model is based on constant reactivity so it cannot predict char burnout during the later stages accurately. Unlike simple kinetic models [5,6], however, the CBK model includes three deactivation mechanisms capable of creating low reactivity at high conversions: thermal annealing, ash inhibition, and preferential consumption of more reactive char.

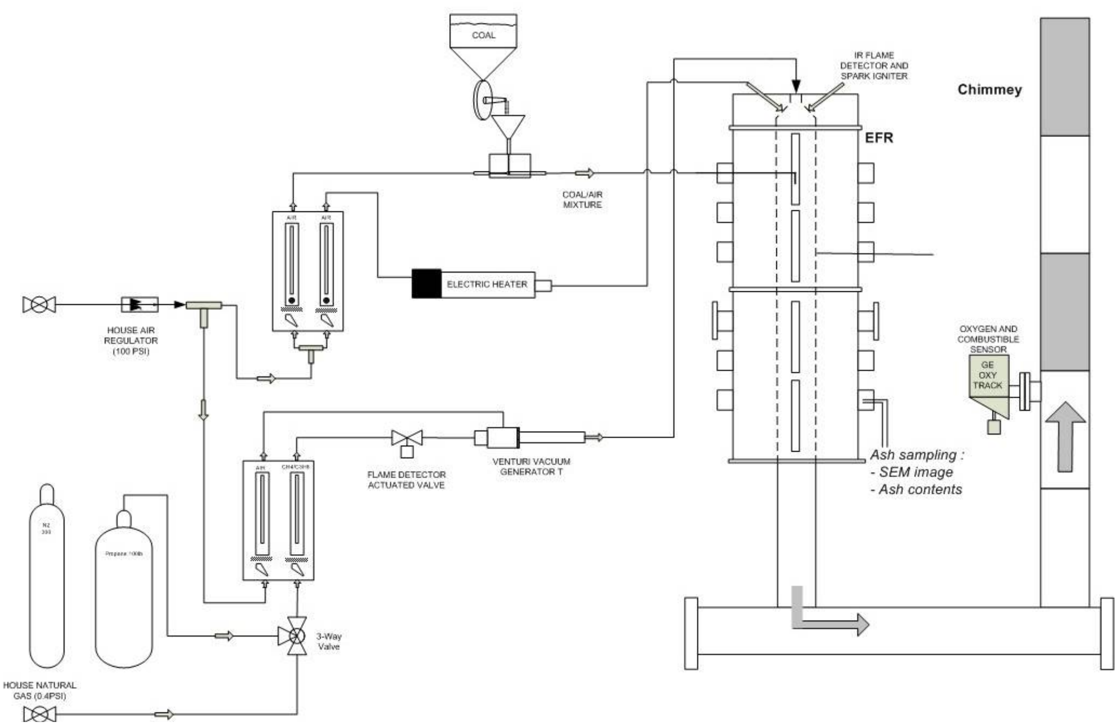

Figure 2 - Schematic Diagram of Entrained Flow Reactor [7]

Experimental method:

The char burnout experiments were performed in a nominally one-dimensional (i.e., changes occur only axially), flame supported entrained flow reactor. The EFR consists of a tube 2.0m long with an internal diameter of 0.2m. In order to control the temperature of the furnace and reduce heat loss, three cast layers of refractory with descending thermal conductivity insulate the EFR wall. The total thickness of the refractory is 0.35m. Propane and air are premixed and are injected uniformly with an equivalence ratio of 0.74. This provides a thermal and oxygen concentration environment similar to that of a full-scale utility boiler. The EFR gas temperature is in the range of 1100 - 1650 K; a downward directed stream of coal particles is injected into the low speed 1 m/s oxidizing gas environment. Figure 2 shows a detailed schematic of the EFR [7].

Eastern bituminous coal is used for this experiment. The coal is fed at a rate of 1.667 $\times 10^{-4}$ kg/s. A volumetric micro feeder was chosen to feed the coal to the EFR. The mechanism that feeds the coal is a rotating cone. The cone rotates at an angular velocity and a small rib on the inside of the cone constantly pushes coal particles up the slope of the cone [3].

The temperature profiles in the reactor were measured with a K type thermocouple both across the furnace diameter and along the length of the furnace at 0.35 m, 0.58 m, 1.00 m, 1.36 m and 1.58 m from the coal injector. Based on timescale and mixing estimates, the oxygen concentration inside the furnace is presumed essentially uniform radially, but it will be measured with a TESTO combustion analyzer. A GE Oxytrak oxygen sensor is permanently mounted to the exhaust stack for measuring oxygen mole fraction and combustible gas mole fraction at the exit. During coal combustion, the oxygen mole fraction varies from 6 percent at the inlet to 3 percent at the exit. The average residence time of particles in the EFR is approximately 2 seconds.

Ash particles were collected with an approximately isokinetic sampling probe and the degree of particle burnout was determined based on loss on ignition (LOI) values. Samples were collected from two different sampling ports. The first sampling point was the fourth port at a distance of 1 meter from the injection location. The second sample was collected from the last port in the furnace at a distance of 1.6 meters from the coal injector. Each port was sampled continuously for two hours. The LOI measurements were repeated three times on each sampled measurement using the FERCo LOI instrument (Fossil Energy Research Corporation, Lake Forest, California) and a micro scale.

Numerical modeling:

A geometry similar to the entrained flow reactor geometry was created and meshed in Ansys Gambit and imported to Fluent to produce a similar gas phase environment and particle history within the designed EFR. A hexagonal-wedge mesh type was used for the model. A grid-independence check has been performed in which the number of grid points in each direction was doubled to ensure the accuracy of the numerical solutions.

In order to solve the mass, momentum and energy conservation equations, boundary conditions were set at all the boundaries. The boundary condition on the inside wall for the velocity field is the no-slip condition. For the outer wall, it was assumed that heat transfers by radiation to the surrounding environment and by free convection, with the ambient air temperature and those of the surroundings set to 300K. There is a small diffuser centered at the top of the furnace where 16 liter/min of air carries coal particles into the furnace. Surface CO_2 formation must be considered for temperatures below 1700 K [8]. In this study, it was considered that heterogeneous char oxidation produces both CO and CO_2. For volatile gas combustion it was assumed that all the volatiles produce CO first, and a two-step reaction was selected for combustion of volatiles in the gas phase.

In this simulation, a 3D pressure based solver is used. The realizable k-epsilon model is chosen because it produces more accurate results for boundary layer and axisymmetric round jet flows than the standard k-epsilon model [9]. As mentioned earlier, the simple built-in kinetic-diffusion limited combustion model and Fluent's internal CBK model were used for char burnout. The convergence criterion for x,y,z velocity, energy, k, epsilon and species concentrations was 10^{-5}.

A Rosin-Rammler distribution with a spread parameter of 1.366, mean diameter of 45.4μm, minimum diameter of 15μm, and maximum diameter of 300μm was used for coal particle input sizes. These values were chosen based on particle size distributions of the coal used in the experiment. The proximate analysis of the coal was 49.56% fixed carbon, 33.31% volatile matter, 9.66% ash and 7.47% moisture. The coal's ultimate analysis was 69.12% carbon, 4.67% hydrogen, 1.43% nitrogen, 1.35% sulfur, 6.3% oxygen and 9.66% ash and 7.47% water. The fixed carbon was set to 84.8 percentage dry ash free (DAF). The coal had an initial density of 1400 kg/m^3 and an initial temperature of 358.15K was assumed. High temperature volatile matter loss is assumed to be 1.6 times greater than the volatile matter assessed by the ASTM standard proximate analysis. The intrinsic reactivity was set to 3560 gm-C/(gm-C)-sec-(mol/m3)^n. The swelling factor is assumed to be 2 [10]. Following simulation, data pertaining to the time, temperature and oxygen concentration history of the EFR model were extracted.

Since environment oxygen and environment temperature of a single particle cannot be extracted simultaneously using standard built-in functionality of Fluent, a user-defined function code was developed to obtain time, diameter, position, mole fraction of oxygen, environment temperature and mass of the coal particle during char burnout.

Results:

The temperature profile across the EFR was measured during coal combustion with a K-type thermocouple. These measurements were used to ensure that the temperature distribution across the EFR matched the model's temperature distribution at the same locations. According to the measurements, the temperature profiles across the EFR were approximately flat. This uniformity is due to mixing and the presence of carbon dioxide and water vapor in the gas mixture which absorb and re-emit thermal energy by radiation. The EFR's temperature profiles and contour plot are presented in Figure 3 and Figure 4, respectively. Measured temperatures match the numerical results extremely well (less than 3% difference) except at two points near the wall. These wall values will be rechecked using a suction pyrometer to avoid possible radiative errors but since no captured particles travel close to the wall these anomalies are not crucial at this point.

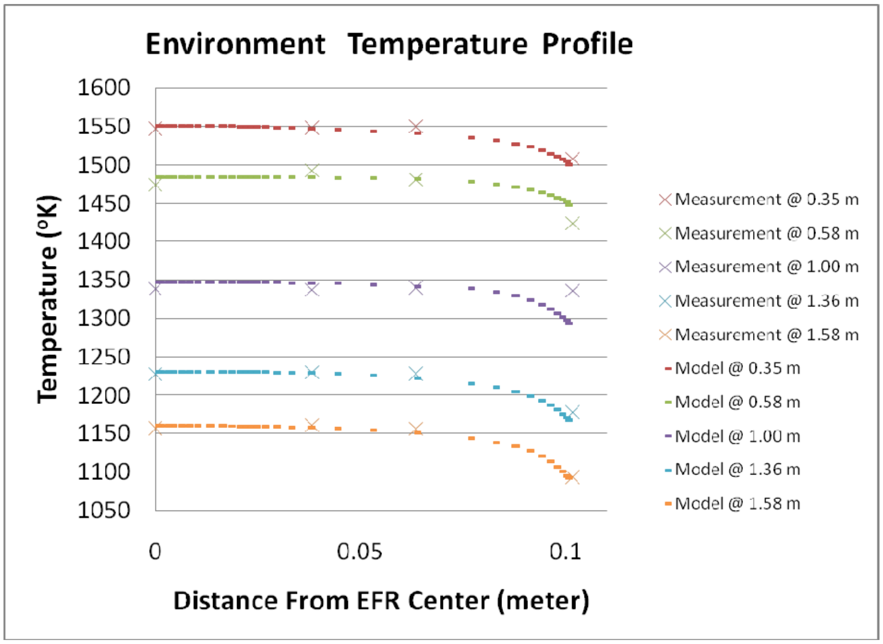

Figure 3 - Temperature profile across the EFR during coal combustion.

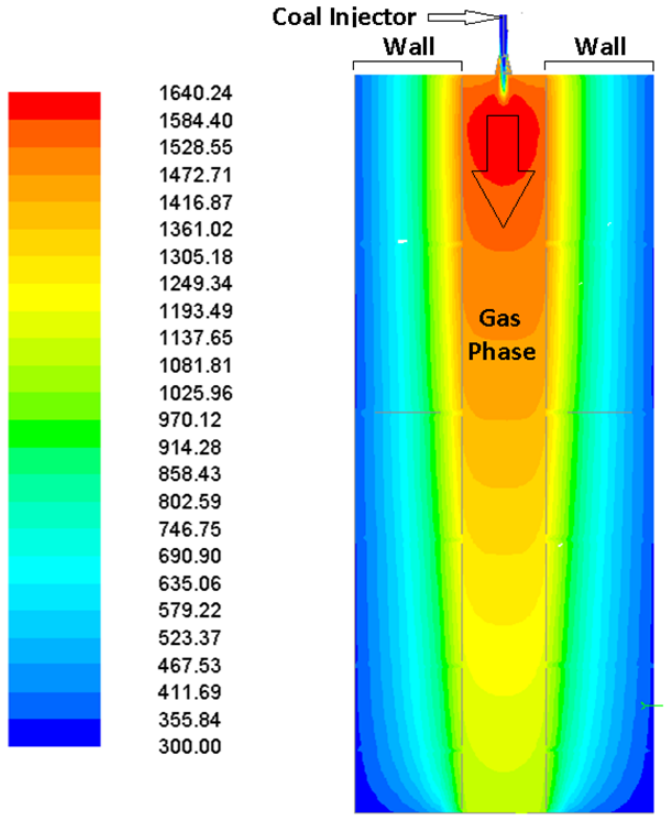

Figure 4 - Fluent simulation predicting static temperature including gas phase and solid phase during Eastern Bituminous coal test.

In order to confirm sufficient accuracy between the model and experiment, a sensitivity analysis was performed to investigate the robustness of the model predictions and assess which input parameter variation most greatly contributes to LOI variability. This analysis allows us to determine what level of accuracy is necessary for environment temperature and oxygen input parameters to make the prediction valid. In this analysis the CBK model was used.

Two 110 micron particles with similar temperature histories were selected. As shown in Figure 5, however, their oxygen-time histories are different by about 15%. Figure 6 shows that this difference leads to an associated 10 percent difference in LOI.

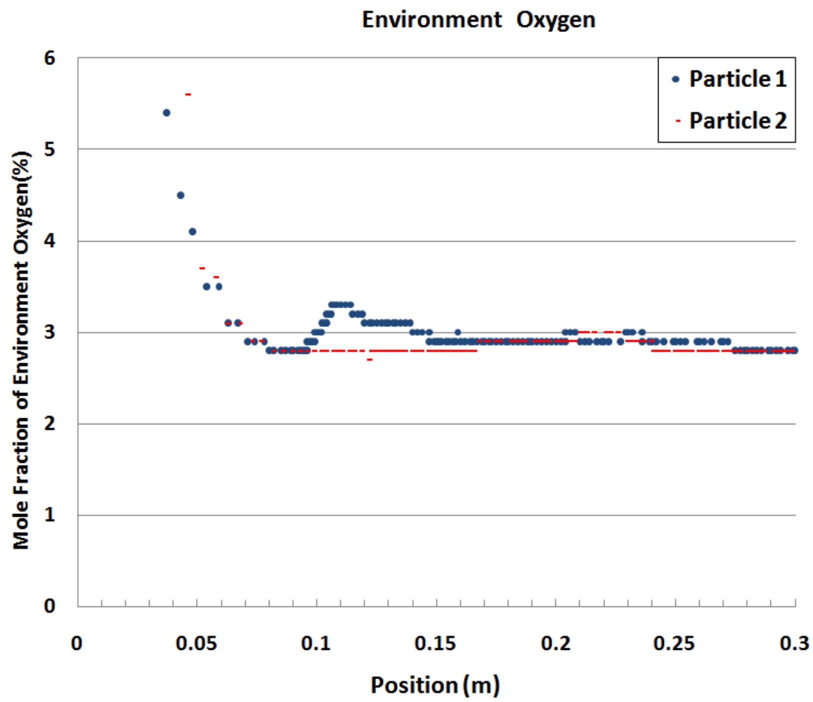

Figure 5 - Oxygen concentration paths of two 110 um particles with similar temperature

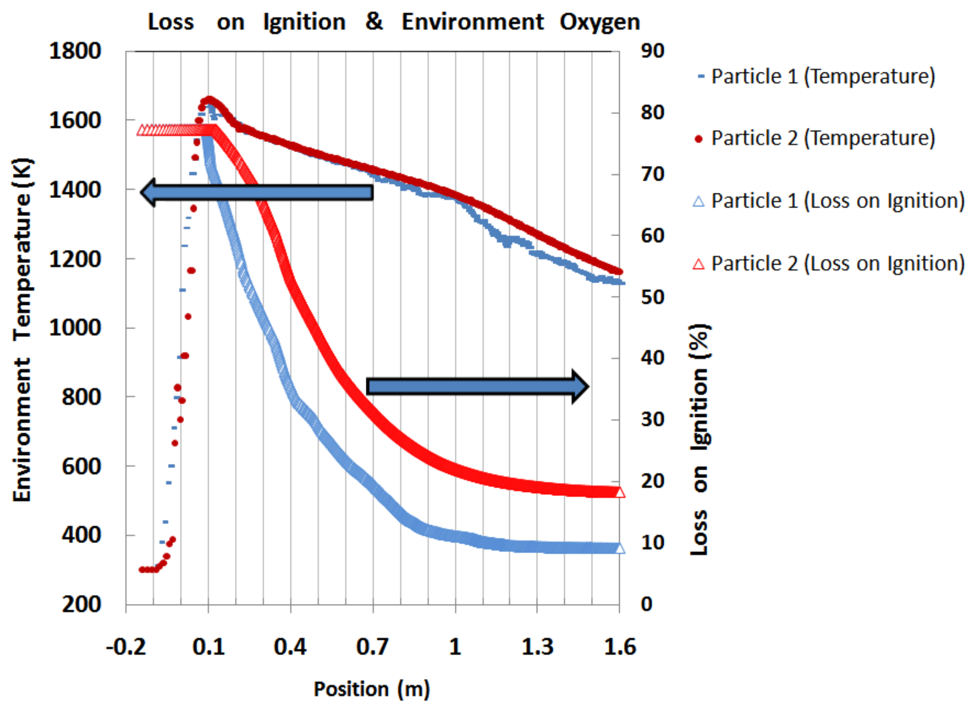

Figure 6 - Environment temperature history and loss on Ignition computed by Fluent embedded CBK model for two 110um.

In order to explore how sensitive the CBK model is to changes in the environment temperature, two 110 micron particles with similar oxygen history were selected (Figure 7). Particle 1 is exposed to 80 K higher temperature relative to particle 2. (Figure 8) The LOI result shows that the 80 K difference in temperature can create a 10% change in LOI (Figure 8).

Therefore, both temperature variation and oxygen variation contribute to substantial LOI effects and it is necessary to measure the environmental oxygen at different locations in the EFR to adjust and validate the EFR model.

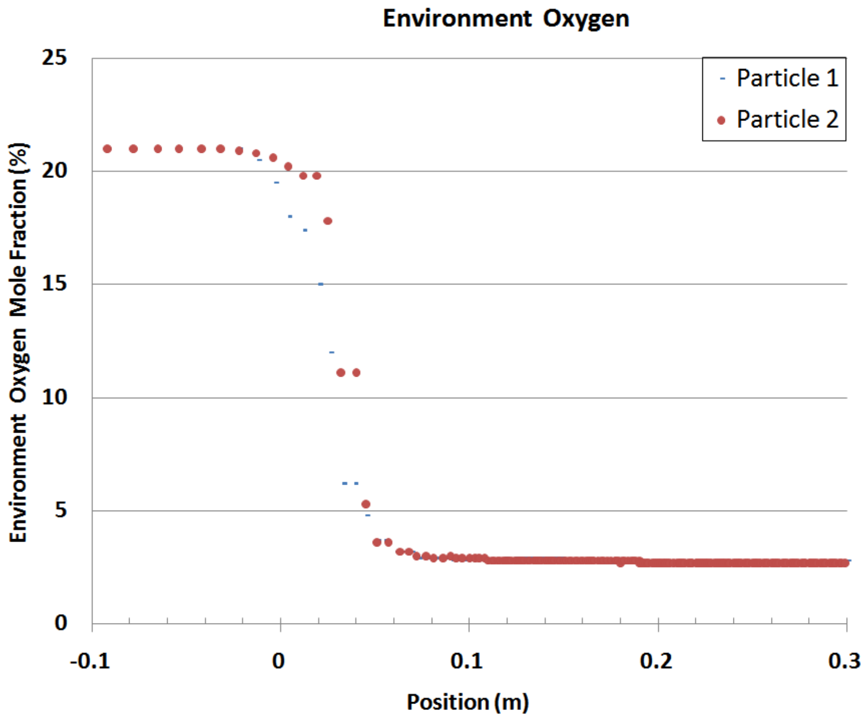

Figure 7 - Oxygen concentration history of two 110um particles with similar oxygen history

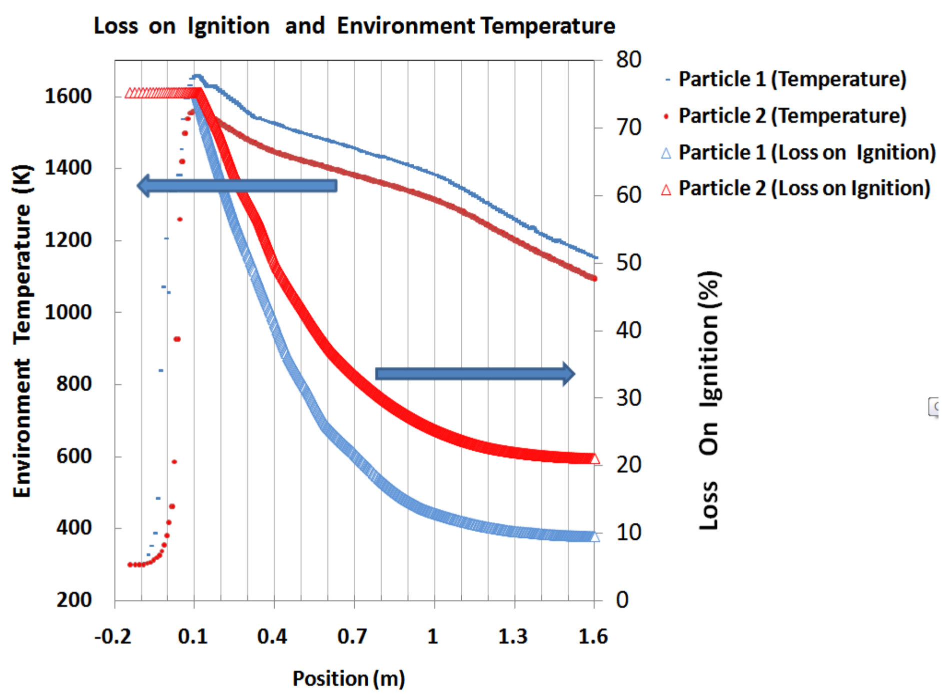

Figure 8 - Temperature history and % Loss on Ignition computed by CBK for two 110um particles

As a demonstration of the project approach, the measured LOI at two different locations,1.0 m and 1.6 m, are shown in Figure 9. Although the oxygen distribution inside the EFR has not been validated, the LOI measurements are compared with the CFD prediction using the CBK model under the same environment temperature condition and the calculated oxygen concentration at the inlet and the exit (Figure 9). The average LOI predicted by CBK and the standard deviation of all 110 micron coal particles are presented in Figure 9. The results show that the non-uniform oxygen distribution in the model causes particles to be exposed to different oxygen histories. According to the sensitivity analysis, it is expected that radial variation in oxygen concentration leads to variation in LOI prediction. However, results show that the average LOI predicted by Fluent embedded CBK for all 110 micron coal particles is in good agreement with the LOI measurements.

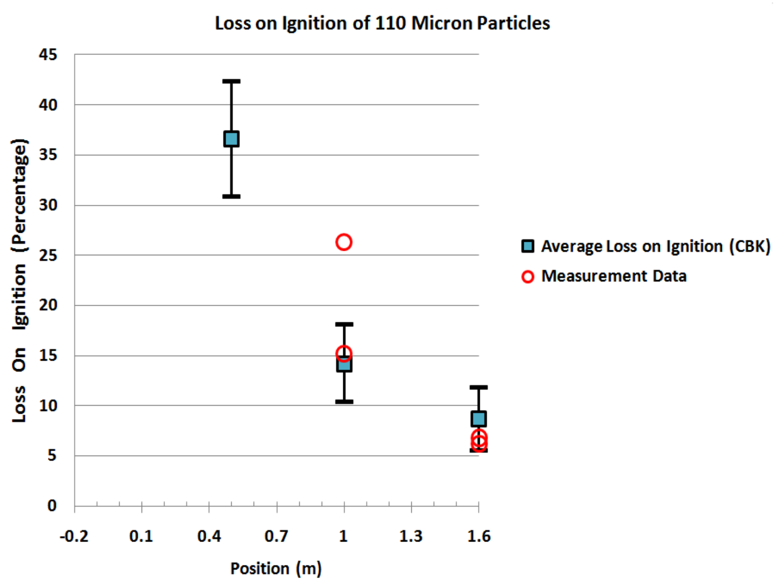

Figure 9 - Measured Loss on Ignition and average Loss on Ignition predicted by CBK model for 110 micron particles

Conclusion:

The objective of this study is to improve models that allow prediction of carbon burnout in the late stages of coal combustion. A numerical model of coal burning in the UC Irvine entrained flow reactor (EFR) was created to evaluate common char burnout kinetic modeling approaches. The measured temperatures match the numerical heat transfer model very well. Based on sensitivity analysis, variation in oxygen and temperature both play important roles in loss on ignition prediction equally. LOI measurements at two different locations were compared with the LOI predicted by Fluent embedded CBK model.

Work in progress :

Efforts currently underway to further validate our modeling results include:

- Oxygen concentration measurements at different axial and radial locations in the EFR.
- Suction pyrometer temperature measurement to reduce the error due to radiation effects.
- LOI measurements for different coals will be conducted under these better characterized environment conditions. These new measurements will be compared to CFD predictions using two standard char burnout sub-models in Fluent – the kinetic-diffusion and CBK, and an external CBK code under the same environmental condition as the EFR.

Acknowledgment:

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