Effect of Heating Rate on the Thermodynamic Properties of Pulverized Coal

**Semi-Annual Report
September 24, 1997 - March 31, 1998**

**By
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For
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EFFECT OF HEATING RATE ON THE THERMODYNAMIC PROPERTIES OF
PULVERIZED COAL

Semi-Annual Progress Report

For the Period September 24, 1997 to March 23, 1998

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ABSTRACT

This semi-annual technical progress report describes work performed under DOE Grant No. DE-FG22-96PC96224 during the period September 24, 1997 to April 23, 1998 which covers the third six months of the project. During this reporting period, several components of the electrodynamic balance measurement system, Single Particle Laboratory, Federal Energy Technology Center, Morgantown, WV, were successfully calibrated. A large number of single polystyrene spheres covering a size range of 80 - 200 μm in diameter were caught in the electrodynamic balance. The size counts of their projected images obtained using the top video-based imaging system, bottom video-based imaging system, and diode-array imaging system were calibrated against the actual size of the particles to within $\pm 3 \mu\text{m}$. Signals obtained by the particle position control system were also calibrated against the actual movement of a polystyrene particle in the balance to within $\pm 1 \mu\text{m}$. Presently, calibration of the Single Color Pyrometer to measure coal particle temperature histories is in progress. Donation agreement for the Heated-Grid measurement system from our industrial partner, United Technologies Research Center (UTRC), CT, was obtained and the arrangement for the completion of the shipment of the grid system components from UTRC to CAU is in progress. Several theoretical analyses were conducted to improve the model performance of the present work and the results were compared with data available from our previous studies. These activities resulted in several publications including three conference papers, and one student poster paper during this reporting period.

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EXECUTIVE SUMMARY

In this semi-annual report, the work performed under DOE Grant No. DE-FG22-96PC96224 during the period September 24, 1997 to April 23, 1998 is described and the major accomplishments are highlighted summarizing the most important research results.

In all major coal conversion processes, coal undergoes a devolatilization stage while it is heated to the reaction temperature. The recent experimental studies of devolatilization of pulverized coal at rapid heating rates representative of coal combustors have greatly improved our general understanding of this process. But the heat transfer analysis with commonly-applied thermal properties developed from slow heating rate experiments did not predict either the early heating or the latter stages of heating. Knowledge of the role of heating rate on coal thermal properties is essential to progress in advanced coal utilization technology. The objectives of this proposal are to understand the effect of heating rate on thermal properties of pulverized coal particles. The specific objectives are to subject coal particles into a broad range of heating rates, measure temperature histories, and develop thermal property (heat capacity and thermal conductivity) data-base for use in advanced coal combustion modeling. Experiments and modeling are being carried out to meet the project objectives. The successful accomplishment of the above goals will provide better understanding and unique predictive capability for coal thermal properties for use in high heating rate applications.

During this reporting period, several components of the electrodynamic balance measurement system at the Single Particle Laboratory, Federal Energy Technology Center, Morgantown were successfully calibrated. Single polystyrene spheres were caught in the electrodynamic balance and the size counts of their projected images obtained using the top video-based imaging system, bottom video-based imaging system, and diode-array imaging system were calibrated against the actual size of the particles. The polystyrene spheres were retrieved after each experiment and sized under a microscope to within $\pm 3 \mu\text{m}$. A polystyrene sphere stuck in a glass plate was suspended in the balance by a clamp attached to an extended pole mounted on a XYZ translation stage. The position signals obtained using the particle position control system was calibrated to within $\pm 1 \mu\text{m}$ to the actual movement of the particles measured by the XYZ stage. Presently, calibration of the Single Color Pyrometer to measure coal particle temperature histories is in progress. Donation agreement for the Heated-Grid measurement system from our industrial partner, United Technologies Research Center (UTRC), CT, was obtained and the arrangement for the completion of the shipment from UTRC to CAU is in progress. Following the complete arrival of the donated components of the Heated-Grid system, installation, testing, and low heating rate devolatilization experiments will be performed employing this system at CAU. Several theoretical analyses were conducted to improve the model performance of the present work and the results were compared with data available from our previous studies. These include analysis of surface area and volume for irregularly-shaped coal particles, sensitivity analyses on heat capacity and thermal conductivity for coals, and assessment of kinetic parameters for coals during devolatilization. These activities resulted in several publications including three conference papers, and one student poster paper during this reporting period.

INTRODUCTION

This semi-annual technical progress report describes work performed under DOE Grant No. DE-FG22-96PC96224 during the period September 24, 1997 to April 23, 1998 which covers the third six months of the project.

In all major coal conversion processes, coal undergoes a devolatilization stage while it is heated to the reaction temperature. Recent experimental studies of devolatilization of pulverized coal at rapid heating rates representative of coal combustors have greatly improved our general understanding of this process [1-18]. But the heat transfer analysis with commonly-applied thermal properties developed from slow heating rate experiments did not predict either the early heating or the latter stages of heating [1-3,18-19]. Design of coal combustion and conversion processes require knowledge of thermal properties to construct an energy balance. It is accepted that there are uncertainties in the heat capacity of coal especially for the high heating rate studies [2,6]. It is also accepted that the large thermal gradients within the particle (due to thermal conductivity of coal) make prediction of the temperature difficult during the early heating in these studies [6]. However, there has been no independent study conducted to investigate the effect of heating rate on the thermal properties of coal particles. Knowledge of the role of heating rate on coal thermal properties is essential to progress in advanced coal utilization technology.

The objectives of this proposal are to understand the effect of heating rate on thermal properties of pulverized coal particles. The specific objectives are:

- 1) Subject coal particles into a broad range of heating rates and extract heat capacity information for high heating rate applications.
- 2) Also develop thermal conductivity information for coal subjected to rapid heating rates representative of coal combustors.

Experiments and modeling are being carried out to meet the project objectives. The successful accomplishment of the above goals will provide better understanding and unique data base for coal thermal properties for use in high heating rate applications.

PROGRESS TO DATE

Literature articles [1-45] were reviewed to understand the effect of heating rate on various parameters of a coal particle. The role of heating rate on the onset of volatile evolution, volatile yield, product composition to a lesser extent, coal type and particle size were found to be well established. As heating becomes more rapid, the onset of devolatilization shifts to much smaller time scales and to much higher surface temperatures [2,18-19]. However, the role of heating rate on coal thermal properties was not found to be well understood. Previous results clearly demonstrated that particle temperature-dependent thermal property assumptions routinely applied in coal combustion models result in large errors (up to 100 percent) in calculated temperature histories [2,18-19].

Remote access from CAU to the VAX/VMS system at FETC/Morgantown was established through telnet. Several modifications to the existing Single Particle Heat Transfer code were made that include option for various input and/or combinations of input for thermal properties such as constant, temperature-dependent, heating rate dependent, and time dependent values. Many sensitivity analyzes were employed to evaluate and quantify the impact of several heat transfer parameters for coal such as thermal properties, geometry related properties such as shape, mass, and density, and chemical properties such as devolatilization kinetic coefficients on rapid heating. This work improved the model performance of the present work, and also aided in several publications.

Approval for physical access to the Single Particle Laboratory at FETC/Morgantown from Morgantown facility was obtained. This required completion of several laboratory safety courses at the Morgantown site. Several coal particles were caught in the electrodynamic balance system at FETC/Morgantown and the operation of the associated system components such as video-based imaging systems, diode-array imaging system, dc and ac power supply, particle position control, and GE tungsten strip lamp & lens systems for pyrometer were tested and found to be performing well. Testing of the Single Color Pyrometer to measure coal particle temperature histories was completed and found to be satisfactory. Recently, completed calibration of several components of the balance measurement system including imaging systems and particle position control system.

Approvals from FETC/Pittsburgh, CAU, and UTRC for performing the heated grid experiments at Clark Atlanta University (CAU), GA, that were originally planned at United Technologies Research Center (UTRC), CT, were obtained and a partial shipment of the heated grid system components from UTRC to CAU was received. More recently, donation agreement for the Heated-Grid measurement system from UTRC was obtained and the arrangement for the shipment of the remaining components from UTRC to CAU is in progress. Purchase of several instrumentation that were not available in the package from UTRC that include power supply, data acquisition interface, and spot welder was made by CAU.

WORK PERFORMED DURING THIS PERIOD

During this reporting period, several components of the electrodynamic balance measurement system, Single Particle Laboratory, Federal Energy Technology Center, Morgantown, WV, were successfully calibrated.

A large number of single polystyrene spheres covering a size range of 80 - 200 μm in diameter were caught in the electrodynamic balance. Single particles were backlit with a red He:Ne laser at the side and with a light emitting diode (LED) from the bottom of the balance. The magnified shadow image of the side view was split and projected onto the detector of a CCD video camera imaging system and a high-speed diode array imaging system. The magnified shadow image of the bottom view was projected onto the detector of a second CCD video camera imaging system positioned above the balance. The size counts of the projected images obtained using the top video-based imaging system, bottom video-based imaging system, and diode-array imaging system were calibrated against the actual size of the particles to within $\pm 3 \mu\text{m}$. The polystyrene spheres were retrieved after each experiment and sized under a microscope. The video-based imaging systems (side and top) are

used for routine particle observation and shape characterization. The horizontal counts and the vertical lines blocked by the projected images were recorded in both cameras. The total number of horizontal counts blocked was proportional to the cross-sectional area of the particle. The total number of vertical lines blocked was proportional to the diameter of the particle. The total number of counts in the largest line was also found proportional to the diameter of the particle.

The diode array imaging system is used to characterize particle shape, position, and drag coefficient/mass (C_d/m) ratios. It is composed of 16 x 62 elements of silicon photodiodes spaced 100 Fm apart. The magnified image of the particle projected onto the array blocked a certain number of photodiode elements yielding an analog output (at 6200 Hz) that was found proportional to the cross-sectional area of the particle in the EDB.

A polystyrene sphere stucked in a glass plate was suspended in the balance by a clamp attached to an extended pole mounted on a XYZ translation stage. The position signals obtained using the particle position control system was calibrated to within $\pm 1 \mu\text{m}$ to the actual movement of the particles measured by the XYZ stage.

Presently, calibration of the Single Color Pyrometer to measure coal particle temperature histories is in progress. Measurements of the radiant emissive power from the heated particles will be made using the pyrometer focussed on the same optical access port as the diode array. The pyrometer incorporates a thermoelectrically cooled InGaAs detector. The detector is filtered to provide a 100-nm bandpass centered at 1.5 Fm with high blocking levels at the HeNe ($> 10^{-5}$) and Nd:YAG ($> 10^{-9}$) wavelengths. The pyrometer is being calibrated against a standard General Electric tungsten strip lamp to a temperature greater than 1500 K. This involves imaging the lamp filament with a lens system onto a pinhole of known size at the balance center. The radiant energy from the pinhole will then be collected and re-imaged onto the pyrometer detector with the use of a second similar lens system. An effective wavelength of 1.5 Fm will be filtered to reproduce lamp temperature data within $\pm 3 \text{ K}$. The operation of the GE lamp and its associated optics to focus the lamp emission into the particle location were tested and found satisfactory.

Dr. Sampath, PI of this project, was at FETC/Morgantown working in this project for couple of weeks during this reporting period. He was involved in the alignment and setting up of the pyrometry system. Dr. Esmail Monazam who has coauthored with the PI in a number of publications in the subject matter is working as a part-time research associate in this project and is stationed at FETC/Morgantown. Dr. Monazam is involved in the experimental activities discussed above.

Donation agreement for the Heated-Grid measurement system from our industrial partner, United Technologies Research Center (UTRC), CT, was obtained by CAU and the arrangement for the completion of the shipment of the grid system components from UTRC to CAU is in progress. Following its arrival at CAU, installation, testing, and coal heating experiments will be conducted at CAU employing the heated-grid system that were originally planned at UTRC.

Joie C. Taylor, an undergraduate student in Engineering, is supported and trained in the subject matter. Joie is presently involved in numerical modeling, and spot welding of the

thermocouples on the surface of steel wire-mesh to measure temperatures in the heated grid experiments. Several theoretical analyses were conducted to improve the model performance of the present work and the results were compared with data available from our previous studies. These activities resulted in several publications including three conference papers, and one student poster paper during this reporting period.

Simulations using temperature dependent heat capacity and thermal conductivity correlations routinely applied to coal severely under predicted the particle temperature rise during the early stages of heating. Sensitivity analyses were performed and it was found that the constant room temperature values for heat capacity and thermal conductivity showed excellent agreement with measurements prior to devolatilization during rapid heating. Increases in heat capacity and thermal conductivity observed under slow heating conditions result from bond breaking and structural changes which lead to an increase in vibrational modes of freedom in the coal structure. This suggests that under rapid heating conditions the coal structure is frozen and that these vibrational modes only become accessible at higher temperatures or longer soak times. Two heat capacity curves were used independently in the analysis to model temperature histories during devolatilization: one for the parent coal with the room temperature value and a second for the excited lost volatile molecules with the temperature-dependent values. Model predictions agreed well with data available in our previous studies. A paper and a student poster discussing these results were presented at the Sixth Annual DOE/HBCU/MI/Industry Technology Transfer symposium, April 27-29, 1998, Ocean City, MD. Joie was presenting the poster paper.

A simple analysis following the approach of Heywood [50] was developed to predict surface area and volume for irregularly shaped particles. Predicted results for 21 coal particles were compared with the actual measurements of surface area and volume equivalent diameters available in our previous studies. It was found that the simple analysis predicts the measurements well to within $\pm 4\%$. This finding is useful for the treatment of shape for irregularly-shaped coal particles in heat transfer calculations and will be presented in a paper at the Central States Technical Meeting of the Combustion Institute, June 01, 1998, Lexington, KY.

Devolatilization temperature histories for coal particles heated with high heating rates on the order of 10^4 - 10^5 K/s were modeled successfully with a detailed heat transfer analysis that accounted for coupled chemical, mass, and thermal transport processes occurring during devolatilization. High speed films taken during the experiments in our previous studies provided a clear indication of the time-scales for the commencement and duration associated with volatile evolution. Accurate modeling of temperature history using the above approach required some understanding of mass loss kinetics because it is tied to volatile evolution. Since a direct measure of mass loss was not possible in the study associated with electrodynamic balance system, volatile evolution time-scales were used to infer mass loss kinetics. Data for these observed time-scales and corresponding temperatures for the commencement and completion of volatile evolution for about 40 hva bituminous coal particles tested previously were compared with the calculated time-scales and temperatures using several rate parameters (fast, medium, and slow) found in the literature. The first-order kinetic equation for the medium rate parameters was derived and found to predict the measurements to within experimental error. A paper summarizing these results will be presented at the 1998 ASME International Energy

Systems conference, September 20-23, 1998, Atlanta, GA. Possible reasons for the poor prediction by the other rate parameters under the conditions employed in this analysis will also be discussed.

OUTCOMES TO DATE

1. Sampath, R., Monazam, E. R., Maloney, D. J., and Zondlo, J. W., Development of Improved Coal Combustion Modeling: Analysis of Coal Particle Irregularity and Thermal Properties on Temperature Predictions, Fifth Annual HBCU conference organized by FETC/DOE at Southern University and A&M College, LA, March 1997.
2. Sampath, R., Maloney, D. J., and Monazam, E. R., Effect of Heating Rate on the Thermodynamic Properties of Pulverized Coal, Contractors Review Meeting, Fifth Annual HBCU conference organized by FETC/DOE at Southern University and A&M College, LA, March 1997.
3. Taylor, J. C., Sampath, R., Maloney, D. J., Zondlo, J. W., and Monazam, E. R., Transport Phenomena of Irregularly-Shaped Solid Particles in an Electrodynamic Balance, Poster Paper, Fifth Annual HBCU conference organized by FETC/DOE at Southern University and A&M College, LA, March 1997.
4. Sampath, R., Maloney, D. J., Zondlo, J. W., and Monazam, E. R., Temperature Histories for Single Coal Particles Prior to Devolatilization, Central States Technical Meeting, Combustion Institute, April 1997, Point Clear, AL.
5. Taylor, J. C., Sampath, R., and Maloney, D. J., Characterization of Small Single Particles in an Electrodynamic Balance, Annual Student Scientific Research Symposium, April 15, 1997, Clark Atlanta University, Atlanta, GA.
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11. Sampath, R., Maloney, D. J., and Zondlo, J. W., Measurements of Surface Area and Volume for Irregularly-Shaped Coal Particles, Central States Technical Meeting, Combustion Institute, 31 May - 2 June, 1998, Lexington, KY.
12. Sampath, R., Maloney, D. J., and Zondlo, J. W., Analysis of Devolatilization Time-Scales, Temperature Histories, and Kinetics for Coal Particles Subjected to Rapid Heating, Computers in Energy Systems, 1998 Eighteenth ASME International Computers in Engineering Conference, Atlanta, GA, September 20-23, 1998.

PLANS FOR THE NEXT REPORTING PERIOD

In the near future, pyrometer calibration will be completed at FETC, Morgantown, and Heated-Grid reactor will be installed at the Research Science Center, Clark Atlanta University. Following these activities, both the systems will be tested for temperature measurements. Heating experiments will be performed on coal particles, and temperature data collected for a broad range of heating rates.

SUMMARY AND CONCLUSIONS

The project is progressing well. Calibration activities for various instruments of the EDB measurement system, Single Particle Laboratory, FETC, Morgantown, were initiated, and 90% completed. Calibration activities are expected to complete shortly. Donation agreement for the donation of several components of the heated-grid system from UTRC, our industrial collaborator in this project, was obtained. Transit of equipment from UTRC to CAU is in progress. Following its arrival at CAU, installation, testing, and coal heating experiments will be conducted at CAU employing the heated-grid system that were originally planned at UTRC. Modification of the Single Particle Heat Transfer code to include effect of heating rates on predictions was complete and several sensitivity analyses were performed to understand the effect of various key transport parameters involved in the model and to improve the model performance of the present work. An undergraduate student is trained in the subject matter. These activities resulted in several papers, including a student poster paper, for presentation at national and international conferences.

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