SECTION I. INTRODUCTION

I.A. PROGRAM BACKGROUND AND DESCRIPTION

During the past 5 years, significant advances have been made at Brigham Young University (BYU) in comprehensive two-dimensional computer codes for mechanistic modeling of entrained-bed gasification and pulverized coal combustion. During the same time period, significant advances have been made at Advanced Fuel Research, Inc. (AFR) in the mechanisms and kinetics of coal pyrolysis and secondary reactions of pyrolysis products. The proposed program presents a unique opportunity to merge the technology developed by each organization to provide detailed predictive capability for advanced coal conversion processes. This predictive capability will incorporate advanced coal characterization techniques in conjunction with comprehensive computer models to provide accurate process simulations.

The program will streamline submodels existing or under development for coal pyrolysis chemistry, volatile secondary reactions, tar formation, soot formation, char reactivity, and SO_x-NO_x pollutant formation. Submodels for coal viscosity, agglomeration, tar/char secondary reactions, sulfur capture, and ash physics and chemistry would be developed or adapted. The submodels would first be incorporated into the BYU entrained-bed gasification code and subsequently, into a fixed-bed gasification code (to be selected and adapted). These codes would be validated by comparison with small scale laboratory and PDU-scale experiments. The validated code could then be employed to simulate and to develop advanced coal conversion reactors of interest to METC.

I.B. OBJECTIVES

The objectives of this proposed study are to establish the mechanisms and rates of basic steps in coal conversion processes, to integrate and incorporate this information into comprehensive computer models for coal conversion processes, to evaluate these models and to apply them to gasification, mild gasification and combustion in heat engines.

I.C. APPROACH

This program will be a closely integrated, cooperative effort between AFR and BYU. The program will consist of four tasks: 1) Preparation of Research Plans, 2)

Submodel Development and Evaluation, 3) Comprehensive Model Development and Evaluation, and 4) Applications and Implementation.

I.D. CRITICAL TECHNICAL ISSUES

To achieve the goals of the program, the computer models must provide accurate and reliable descriptions of coal conversion processes. This will require the reduction of very complicated and interrelated physical and chemical phenomena to mathematical descriptions and subsequently to operational computer codes. To accomplish this objective a number of technical issues must be addressed as noted below.

I	Separation of Rates for Chemical Reaction, Heat Transfer, and Mass Transfer
I	Particle Temperature Measurements Using FT-IR E/T Spectroscopy
I	Functional Group Description of Coal, Char, and Tar
I	Tar Formation Mechanisms
I	Char Formation Mechanisms
I	Intraparticle Transport
I	Pyrolysis of Volatiles and Soot Formation
I	Secondary Reaction of Tar
I	Particle Ignition
I	Ash Chemistry and Physics
I	Particle Optical Properties
I	Code Efficiency and Compatibility for Submodels
I	Coupling of Submodels with Comprehensive Codes
I	Comprehensive Code Efficiency
I	Turbulence
I	SO_x and NO_x
●	Generalized Fuels Model
I	Fixed-Bed Model

(●) to be addressed; (I) initiated; (C) completed.

These technical issues are addressed in the three Tasks as described in Section II-IV.

I.E. SUMMARY OF SEVENTH QUARTER PROGRESS

Subtask 2.a. Coal to Char Chemistry Submodel Development and Evaluation

Additional improvements were made in the FG-DVC model. A second version of the model was developed for reactors in which the tar is quenched after being evolved. The model run procedure was modified so that successive cases could be run in a batch mode and the results could be averaged, stored and/or plotted. The creation and loss of methyl groups in the DVC model due to bridge breaking and in the FG model due to methane formation was made consistent. In addition, improvements were made in the way that crosslinking is treated in the model.

The FG-DVC model was used to predict baseline pyrolysis data for the eight Argonne coals from three different reactors. In general, the model did a good job in predicting the data for gas, tar and char yields and for the tar molecular weight distributions.

In order to refine the combined kinetic/mass transport submodel used in the FG-DVC model, a search was made of literature pyrolysis data for the Pittsburgh Seam coal, starting with heated grid experiments. When a comparison was made of data produced by heating at 1000 K/s to various peak temperatures, it was found that the results of different investigators did not agree, even when obtained from the same laboratory. We begin an experimental and theoretical study into possible reasons for these variations, which we are doing in conjunction with Professor Eric Suuberg of Brown University.

Subtask 2.b. Fundamental High-Pressure Reaction Rate Data

Several char samples were prepared with the simple hot-tube reactor, and a char sample was collected from the BYU gasifier. These samples have been analyzed by a CH analyzer and SEM. Hydrogen content generally decreases as residence time increases. Some samples, however, showed the opposite effect, probably due to a buildup of tar on the walls of the reactor tube which effectively decreased the residence time. Particles with longer residence times were more porous. The gasifier char, however, was non-porous. Construction of the high-pressure, controlled-profile reactor continued.

Subtask 2.c. Secondary Reaction of Pyrolysis Products and Char Burnout

Data collection was temporarily suspended in the TWR as the spectrometer was needed for a different project. Some of the work done previously on this program was summarized in a paper accepted for presentation at the 22nd Symposium (Int) on Combustion in Seattle, Washington (Aug. 1988) which is included as Appendix A.

Subtask 2.d. Ash Physics and Chemistry Submodel

In order to further understand the role played by ion-exchanged cations on char reactivity, samples of demineralized Zap coal were subjected to ion-exchanged experiments with Ca, Na, and K. Ca exhibited a saturation effect, while Na and K exhibited a maximum in reactivity with increased loading.

Subtask 2.e. Large Particle Submodels

The construction of the fixed-bed reactor was nearly completed. Discussions were held with Dr. Radulovic (from BYU) on the relationship between the fixed-bed reactor model and the large particle pyrolysis model.

Subtask 2.g. SO_x/NO_x Submodel Development

Development of the combined thermal- and fuel-NO_x submodel has proceeded with two computer simulations being made to evaluate the theory and code. The first case was for an entrained-flow coal/oxygen gasifier. Predictions for this case were favorable, showing improvement of the predicted NO concentrations from inclusion of thermal NO. The second case was for a propane/air turbulent diffusion flame. This prediction was also favorable, showing that the Zeldovich mechanism, assuming equilibrium of molecular and atomic oxygen, correctly predicts NO concentrations in post-flame regions, provided the temperature and oxygen concentration are correctly predicted.

A thorough review was made of alternative fuel-NO global mechanisms. A kinetic path including ammonia species as an intermediate is being sought to potentially improve prediction of fuel NO formation from low-rank coals. A survey of on-going SO_x research was also continued and the important elements of a SO_x submodel capable of predicting the conversion of fuel-bound sulfur to gas species have been identified.

Subtask 2.h. NO_x/SO_x Submodel Evaluation

The cold-flow facility was modified with a flow straightener to eliminate recirculation and replicate the flow patterns in the aft-region of the gasifier. Tracer gas and smoke visualization tests are in progress. The gasifier was recast and a flash tank was installed. Several checkout tests were conducted, including one with sorbent mixed with the coal. Thermocouples inserted into the reaction chamber to measure gas temperature failed. A new gas-quenched sample system was developed. Improvements were made to the chemical analysis procedures to better analyze the sulfur capture, and pulverized samples of coal and limestone were prepared.

Subtask 3.a. Integration of Advanced Submodels into Entrained-Flow Code, with Evaluation and Documentation

The FG/DVC model was integrated into a more recent version of PCGC-2 and the new version was transferred to AFR. The new code was tested for Pittsburgh seam coal, and FG/DVC submodel predictions were compared with predictions obtained with the two-step devolatilization model using the rate constants of Kobayashi et al. (1977). Development of a graphics interface using the UNIRAS software package was initiated. A review of the FG/DVC submodel was initiated, and potential improvements were identified. The review of advanced devolatilization models continued, and a laminar code was identified for investigating the relative effects of various devolatilization models in the absence of turbulence effects. Plans were initiated for developing a new approach to chemistry/turbulence modeling based on a Lagrangian description of the gas phase.

Subtask 3.b. Comprehensive Fixed-Bed Modeling Review, Development, Evaluation, and Implementation

The computer code development for the improved fixed-bed gasifier model was continued. The development of the advanced fixed-bed gasifier model was initiated. The collection of fixed-bed reactor design and test data was continued. A comprehensive review of fixed-bed combustion and gasification was initiated.

Subtask 4.a. Application of Generalized Pulverized Coal Comprehensive Code

No work was scheduled.

Subtask 4.b. Application of Fixed-Bed Code

No work was scheduled.