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. FIRST YEAR PROGRESS

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

Characterization by FT-IR, TGA and pyrolysis experiments were carried out on most of the Argonne coal samples, both in bulk and in the glass ampoules. A Rosebud subbituminous coal was also characterized. A study was initiated on the effects of minerals on reactivity by demineralization of the samples using acid treatments. Samples in which alkali metals were removed by ion-exchange methods are also being investigated. Work was done on mass transfer effects in consultation with E.M. Suuberg. The literature search on transport properties during pyrolysis was completed and presented in the Second Quarterly Report. To model pyrolysis data for low heating rate experiments, the internal mass transfer resistance to tar escape must be included. A simple model of tar transport was developed and successfully tested. The FG and DVC models are being modified so that the chemistry of the bridge breaking and crosslinking in the DVC model is completely consistent with the chemistry of gas formation in the FG model. The combined FG/DVC model has been installed on a Sun workstation and programming has been initiated to graphically present the output data. Work was initiated on computing the optical properties of particles during the coal to char transformations.

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

The interdisciplinary research team has been organized. Appropriate literature has been reviewed in pertinent areas. Existing high pressure reactors have been surveyed, and the high pressure reactor and support systems for this study have been designed. The reactor design allows for pressures up to 27 atm, residence times of 1-1000 ms, optical access, preheating of the gas, and wall temperature control. Initial elements of the experimental test plan for the char oxidation and sulfur capture studies have been developed.

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

Studies of ignition and soot formation were performed in the transparent wall reactor (TWR). Several questions on the FT-IR technique were addressed. Attention is being focused on what controls ignition (heterogeneous or homogeneous oxidation) and what controls soot formation. Four samples have been completed: a raw Rosebud subbituminous coal, chars prepared from this coal at 900 and 1500°C and

a demineralized Rosebud coal. To provide data on the relative rate of volatile loss vs oxidation, weight loss measurements are being made in a TGA.

Subtask 2.d. Ash Physics and Chemistry Submodel

The literature search was completed to identify the important effects of coal minerals in determining · al behavior. Characterization experiments were begun on the examination of tne mineral composition of the eight Argonne premium coal samples and one Montana Rosebud subbituminous coal. Work was also initiated to study the mineral transformation during coal conversion.

Subtask 2.e. Large Particle Submodels

A literature review of heat and mass transport effects in coal pyrolysis was completed. In addition, calculations were done to define regimes of internal and external heat and mass transport control for conditions of interest. This was done to define the boundary regions where such considerations become important.

Subtask 2.g. SO_x-NO_x Submodel Development

The existing NO_x submodel in PCGC-2 was evaluated by comparing model predictions with experimental data obtained at high pressure and under fuel-rich conditions. The model gave accurate predictions for bituminous coals but not for lignite. A kinetic model for predicting formation of thermal NO was developed from the literature and coding changes were initiated to incorporate this model into PCGC-2. A literature search of sulfur chemistry in combustion flames was initiated.

Subtask 2.h. NO_x/SO_x Submodel Evaluation

Graduate research assistants have been hired, the experimental program defined, and fabrication of the modified test facility initiated. A cold-flow facility has been designed to simulate the gasifier, providing optical access for laser measurements and flow visualization. A device has been designed to produce smoke so tnat the flow can be visually observed. A method of extracting gas samples from the flow, with on-line sampling and minimal flow disturbance, has been designed so that mixing rate data can be collected.

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

A doctoral student was recruited and began work on this project. Three alternatives are being considered concurrently to integrate the Functional Group (FG) devolatilization model into the comprehensive entrained-bed code. The first method uses existing theory in the code for a single solids progress variable and assumes constant coal offgas composition and enthalpy. During the first year, PCGC-2 was revised under separate funding to include a generalized devolatilization model capable of incorporating the FG model under this method. The second method extends the theory to include multiple solids progress variables and thus allow for variable composition and enthalpy. The general theory was developed and the code modified to allow the two existing progress variables to test this approach. The sensitivity of the code to chemistry/turbulence interactions and to various thermal parameters affecting devolatilization was investigated. A variable solids heat capacity model was incorporated. The third method for incorporating the FG Model is to be based on a probabilistic Lagrangian description of the gas and allows for variable offgas composition and enthalpy while eliminating the need for convolution of gas properties over the mixture fractions. This method offers significant potential for reducing computational time while increasing code stability and accuracy.

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

A literature review of existing fixed-bed coal gasification codes and experimental data was conducted, and three available 2-D codes were installed and tested. Potential improvements over existing models were identified, and an outline of an improved fixed bed model was developed. The proposed model is to be 2-D, steady-state, with separate gas and particle temperatures. Radiative heat transfer within the bed will be modeled. The gas phase will be treated (at least partially) in local equilibrium, and selected pollutant species will be predicted. A review meeting with external consultants was organized to evaluate the proposed model. Several sets of experimental data were obtained for model evaluation.

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

Installation of PCGC-2 on the AFR Apollo DN 580/T computer was completed to allow testing of performance. The test code took approximately 4 times longer than

expected based on the Apollo specifications. The Apollo was returned and Sun Workstations were ordered. PCGC-2 was installed on the Sun Workstation. The reference case for PCGC-2 takes 3.25 hours to run on a SUN 3/260, this compares to 7 hours on the Apollo DN 580 T.

Subtask 4.b. Application of Fixed-Bed Code

No work was scheduled.