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. The status of each of these tasks is also indicated.

A	Separation of Rates for Chemical Reaction, Heat Transfer, and Mass Transfer
A	Particle Temperature Measurements Using FT-IR E/T Spectroscopy
A	Functional Group Description of Coal, Char, and Tar
A	Tar Formation Mechanisms
I	Char Formation Mechanisms
A	Viscosity/Swelling
A	Intraparticle Transport
I	Pyrolysis of Volatiles and Soot Formation
I	Secondary Reaction of Tar
I	Particle Ignition
A	Char Reactivity
I	Ash Chemistry and Physics
A	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
o	Generalized Fuels Model

I Fixed-Bed Model

(o) to be addressed; (I) initiated; (A) almost completed.

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

I.E. FIFTEENTH QUARTER PROGRESS

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

Work continued on the improvements to the FG-DVC model. A set of rank dependent kinetic parameters has been developed for tar, CH_4, CO, H_2O, and CO_2 for each of the Argonne coals. The predictions of the model are being compared to pyrolysis kinetic data from the TG-FTIR system at four heating rates. For a given coal, it is being assumed that the pre-exponential factors are similar for the evolution of the components of a given species (e.g., CO_2-Extra Loose, CO_2-Loose, and CO_2-Tight), but that the activation energies increase in the order Loose ---> Tight. It is also being assumed that the activation energies for each functional group pool increase in the rank order. This approach has been successfully used for the component pools of the major volatile species (CO, CO_2, H_2O, CH_4) for four of the eight Argonne Premium coals, to date (Utah, Pittsburgh, Upper Freeport, Pocohontas). Work on the remaining coals is in progress.

This complete set of kinetic parameters were tested in both Monte Carlo and the 2-σ percolation theory version of the FG-DVC model. Results for three of the eight coals indicate that only small adjustments are required in the kinetic parameters between the two models.

The kinetic parameters are also being validated indirectly through the application of the fluidity model to the Geissler fluidity data for six of the eight Argonne coals. Comparisons are being made to the NMR data of Lynch obtained at CSIRO on the Argonne premium coals. The changes in the fluid fraction predicted from the fluidity model are in good agreement with the change in hydrogen mobility observed in the NMR experiment. We are rechecking the current version of the model with the latest kinetic parameters against van Krevelen's data for fluidity as a function of heating rate and against the isothermal data of Oxley and Pitt.

Work also continued on the swelling model. By using an adjustable parameter for either the diffusion distance or bubble wall thickness in the model, we were able to obtain good predictions for swelling ratio, porosity, and surface area over a range of pyrolysis heating rates.

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

During the last quarter, significant progress in developing the optical particle imaging system and the reactor collection system was made. The optical stands and brackets were designed and fabricated for the HPCP facility. The time required for size classification of pulverized coal was considerably reduced and the quality of the classification was improved significantly. Sufficient quantities of narrow size ranges of two of the test coals were produced for upcoming char preparation and oxidation tests. Analytical techniques to determine tar composition directly from tar filters were also developed and improved accuracy of char weight loss analysis through the use of four simultaneous tracers was investigated. Previously collected char and tar from Pittsburgh No. 8 bituminous coal was analyzed and compared using the new procedures.

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

Additional work was done on the coal flame experiments in the TWR using the FT-IR Emission/ Transmission Tomography technique. Two cases have now been completed for the Montana Rosebud coal (low velocity and high velocity). A low velocity case for the Pittsburgh Seam coal is about 50% complete.

Subtask 2.d. Ash Physics and Chemistry Submodel

No work was scheduled.

Subtask 2.e. Large Particle Submodels

The work on the AFR fixed-bed reactor (FBR) system continued. A redesign of the experiment has been proposed and approved which will allow better quantitation of the tar yields and independent control of the temperature of the second bed. Assembly has begun on a redesign of the experiment. Samples of large particles for each of the eight premium coals have been ordered from Argonne and should be received by July 1, 1990.

Discussions continued with BYU about the single particle FG-DVC model for use in the fixed bed reactor. For this purpose, AFR is developing an ordinary different equation (ODE) version of the 2-σ percolation FG-DVC model. Additional refinements were done to the model to improve its speed. The code was sent to BYU for integration into the Advanced Fixed-Bed Model. A meeting was scheduled at AFR to discuss the single particle model which will involve members of both teams.

Subtask 2.f. Large Char Particle Oxidation at High Pressure

Work continued on designing a large-particle insert to be placed in the HPCP reactor and evaluating analytical procedures for monitoring the rates of large-particle oxidation. A second set of preliminary oxidation experiments were carried out with large char particles made from Utah Bituminous coal, and mass reactivities were determined. The average value of 0.0358 min^{-1} compares favorably with the value of 0.0475 min^{-1} reported previously. Some problems with temperature measurement were experienced.

Subtask 2.g. SO_x-NO_x Submodel Development

Pollutant predictions for coal combustion and gasification cases were completed to verify the extended and alternative fuel NO mechanisms of the submodel. The ability to predict NO formation during gasification of North Dakota lignite was given special consideration. Joint fuel and thermal NO predictions were also completed for combustion and gasification cases. The revised submodel has been fully integrated into PCGC-2 and a user's guideline has been prepared. Work is in progress to complete a sorbent reactions submodel.

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

Work was nearly completed on modeling the transparent wall reactor for code validation and on developing a graphical, user-friendly interface. After modifying for up-firing, gas buoyancy, and laminarization, reasonable results were achieved for the simulation of the non-reacting flow case and the "fast-flow" Rosebud coal flame. Key flame properties, such as ignition point, burnout profile, and gas and particle temperature, have been reasonably well predicted. Complex flow patterns at the nozzle promote particle dispersion, and have not been adequately resolved with current grid spacing. Code predictions are sensitive to inlet boundary conditions for the coal stream at the nozzle exit, and detailed characterization of this boundary condition is needed. Soot formation seems to correlate with equilibrium concentration of condensed carbon, but decays more slowly than predicted from equilibrium. The energy feedback to particles or the CO_2/CO ratio produced at the particle is not adequately predicted.

A window was added to the graphical user interface (GUI) on the Sun workstation for specifying the composition and temperature of the inlet streams. The thermodynamic input file can now be generated automatically using information in the main data file and a chemical species database. Work was also initiated to apply two graphical programs that have been developed under independent funding, a pre- and a post-processor, to PCGC-2. The preprocessor generates a grid file and the post-processor presents computational results. The format of the grid file used by PCGC-2 differs from that used by the pre-processor, and a program was written for converting files between the two formats. A subroutine was modified and added to PCGC-2 for writing the plotting file needed by the post-processor. Both the pre- and post-processors were applied to the TWR simulation described above.

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

Work continued on reviewing, coding, and validating submodels. To enhance user-friendliness, the input file was rewritten to segregate input parameters for the two-zone and one-dimensional submodels. Also, the fixed-bed code was rewritten in a modular fashion with extensive comment statements. The two-zone submodel was improved to accommodate user-specified burnout. Also, two heat transfer zones were added to the well-mixed model to account for heat loss in the freeboard and heat transfer between solid and gas in the ash zone of the reactor. The ash enthalpy calculation was improved.

The fixed-bed code was evaluated by parametric sensitivity analysis. Sensitivity runs were divided into model options, model parameters and operational parameters. Model options include tar vapor reaction equilibrium, volatile mass transport, char ash layer formation, and combustion product distribution. Model parameters include the solid-to-gas heat transfer correction factor, effective diffusivity, bed-to-wall heat transfer, potential tar forming fraction, functional group composition (coal rank), oxidation and gasification kinetics. Operational parameters include the temperature of the feed gas, reactor pressure, coal mass flow rate, steam mass flow rate, air mass flow rate, proximate ash content of the feed coal, proximate moisture content of the feed coal, particle diameter , and bed void fraction.

Subtask 3.c. Generalized Fuels Feedstock Submodel

Work was initiated on modifying PCGC-2 to allow sorbent injection.

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

This subtask has not been initiated.

Subtask 4.b. Application of Fixed-Bed Code

During the last quarter, work continued on collecting fixed-bed design and test data. The advanced fixed-bed code was applied to seven new test cases, including the dry-ash Lurgi gasifier, the Wellman-Galusha gasifier, the METC gasifier, and the slagging BGC-Lurgi gasifier. A presentation was given at the joint METC/AFR/BYU project review meeting, and discussions were held with AFR concerning code development.

Four different gasifier types were simulated: the high pressure, oxygen-fired Lurgi gasifier; the low pressure, air-fired Wellman-Galusha gasifier; the medium pressure, air-fired METC gasifier; and the high pressure oxygen-fired BGC-Lurgi slagging gasifier. Twelve coal types were simulated, which range from lignite to bituminous.