SECTION III. TASK 3. COMPREHENSIVE MODEL DEVELOPMENT AND EVALUATION

Objectives

The objective of this task is to integrate advanced chemistry and physics submodels into a comprehensive two-dimensional model of entrained-flow reactors (PCGC-2) and to evaluate the model by comparing with data from well-documented experiments. Approaches for the comprehensive modeling of fixed-bed reactors will also be reviewed and evaluated and an initial framework for a comprehensive fixed-bed code will be employed after submission of a detailed test plan (Subtask 3.b).

Task Outline

This task will be performed in three subtasks. The first covering the full 60 months of the program will be devoted to the development of the entrained-bed code. The second subtask for fixed-bed reactors will be divided into two parts. The first part of 12 months will be devoted to reviewing the state-of-the-art in fixed-bed reactors. This will lead to the development of the research plan for fixed-bed reactors. After approval of the research plan, the code development would occupy the remaining 45 months of the program. The third subtask to generalize the entrained-bed code to fuels other than dry pulverized coal would be performed during the last 24 months of the program.

III.A. SUBTASK 3.A.-- INTEGRATION OF ADVANCED SUBMODELS INTO ENTRAINED-FLOW CODE, WITH EVALUATION AND DOCUMENTATION

Senior Investigators -- B. Scott Brewster and L. Douglas Smoot

Brigham Young University
Provo, UT 84602
(801) 378-6240 and (801) 378-4326

Objectives

The objectives of this subtask are 1) to improve an existing 2-dimensional code for entrained coal combustion/gasification to be more generally applicable to a variety of coals by incorporating advanced coal chemistry submodels, advanced numerical methods, and an advanced pollutant submodel for both sulfur and nitrogen species, and 2) to validate the advanced submodels in the comprehensive code. The comprehensive code into which the advanced submodels are to be incorporated is PCGC-2 (Pulverized Coal Gasification and Combustion-2 dimensional).

Accomplishments

Work on this subtask is being accomplished under five components: 1) Evaluation and incorporation of coal reaction submodels into the comprehensive code, 2) incorporation of improved numerical solution methods, 3) incorporation of the SO_x-NO_x submodel developed under Subtask 2.g, 4) implementation of the code on computers, and 5) code evaluation. Progress during the last quarter is described below for each of these components.

Component 1 -- Evaluation and Incorporation of Coal Reaction Submodels

This component is aimed at selecting coal reaction submodels and developing methodology for incorporating them into PCGC-2. Three alternatives are being considered for incorporating the single particle model being developed by AFR under Task 2. The first alternative is direct integration, without modification of the treatment of turbulence-chemistry interactions, but allowing for variability in coal offgas enthalpy. This approach is referred to as the Single Solids Progress Variable (SSPV) Method. The second

alternative is to extend the current treatment of turbulence-chemistry interactions to specifically account for variability in coal offgas composition. This approach is called the Multiple Solids Progress Variable (MSPV) Method. The third alternative is a new approach based on treating the gas phase turbulence in a Lagrangian reference frame with a statistical dispersion model. This approach is referred to as the Statistical Gas Dispersion (SGD) Method. Work was conducted during the last quarter on the first two methods, and progress is outlined below. A coal devolatilization model develcped at The University of Utah (Grant et al., 1988) and based on percolation theory is also being reviewed.

Single Solids Progress Variable (SSPV) Method -- The basic assumption of the SSPV Method is that the coal offgas elemental composition is constant and equal to the composition of the original dry, ash-free coal. Under this method, the evolved chemical species predicted by the FG Model are not individually taken into account. Only the overall weight loss is accounted for. The assumption of constant composition allows the mixing of the offgas with the inlet gases to be tracked with only a single progress variable (mixture fraction).

Turbulence/chemistry interactions are accounted for by integrating local instantaneous gas properties calculated from equilibrium over the probability density functions of the coal and inlet gas mixture fractions. Accounting for variability in the offgas enthalpy requires solving the gas energy equation. During the last quarter, the structure of the existing particle model in the 1987 version of PCGC-2 was reviewed to determine an appropriate interface for integrating the single particle model. This structure is illustrated in Figure III.A-1. The model uses a combined Eulerian and Lagrangian approach. The particle number density is calculated on an Eulerian basis, while a Lagrangian approach is used for calculation of particle trajectories and reactions.

The main driver routine for the particle calculations is EOLP (not shown). EOLP first calls CALCNJ to calculate the particle number density fields (n_j) The number density is then passed to the FLUX routines to calculate radiation fluxes (F_x, F_r, and F_ψ) and to the Lagrangian portion of

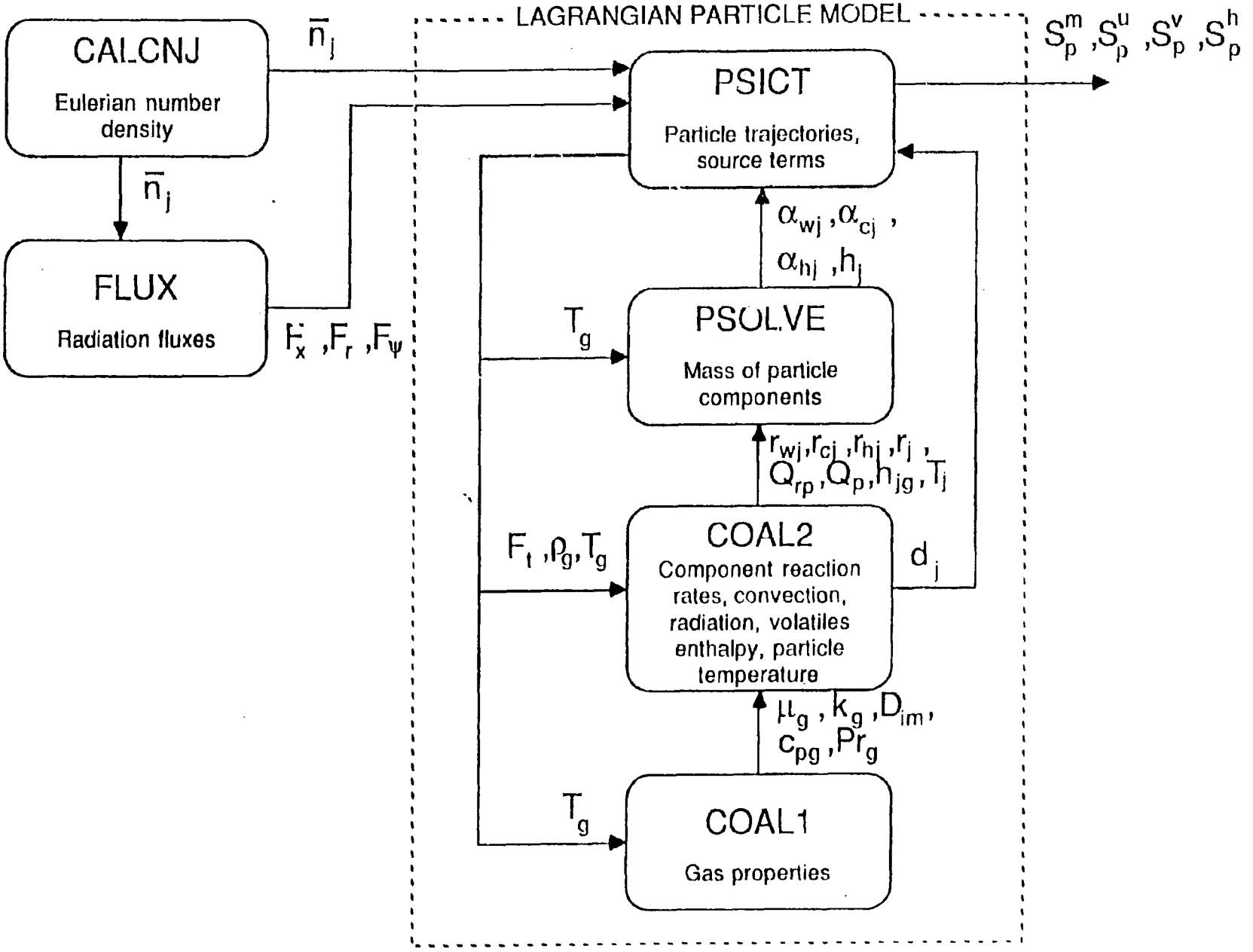


Figure III.A-1. Particle model in 1987 version of PCGC-2.

the particle model. The Lagrangian model consists of four subroutines: PSICT, PSOLVE, COAL2, and COAL1, which calculate the particle trajectories, mass of particle components, particle reaction and heat transfer rates, and local bulk gas properties, respectively. PSICT interpolates the Eulerian gas field to determine local values of the total radiation flux (F_t), gas density (ρ_g), and temperature (T_g). These values are then passed to the other routines. COAL1 uses the local bulk gas temperature to calculate gas viscosity (μ_g), thermal conductivity (k_g), heat capacity (c_{pg}), Prandtl number (Pr_g), and species diffusivity (D_{im}). These values are passed to COAL2. COAL2 calculates particle diameter (d_j) and net rates of change of slurry water (r_{wj}), raw coal (r_{cj}), char (r_{hj}), and total mass (r_j); the rates of radiative (Q_{rp}) and convection (Q_p) heat transfer; the coal offgas enthalpy (h_{jg}); and the particle temperature (Tj). PSOLVE calculates the new particle mass of water ($_{wj}$), raw coal ($_{cj}$), and char ($_{hj}$), and the particle enthalpy (h_j). PSICT keeps track of when particles cross cell boundaries and calculates the source terms to the gas equations for mass (S_m^p), axial velocity (S_u^p), radial velocity (S_p^v), and enthalpy (S_h^p).

The existing particle model in PCGC-2 was found to be lacking in the manner of accounting for heats of reaction. Heats of reaction must be supplied in the input file at the local particle reaction temperature. Since particle temperature varies with time and is calculated independently for each particle size and starting location from the particle energy equation, the reaction temperature is unknown a priori and must be estimated. A more rigorous approach would be to calculate the heats of reaction locally for each time step using heats of formation and heat capacities. These data are already supplied in the main input file for the coal components and in the input file of thermodynamic data for the gas phase components. Changes to implement this more rigorous approach will be made before investigating the effects of changing coal offgas enthalpy.

The appropriate interface for the single particle model is still under consideration. The currently proposed interface is shown in Figure III.A-2. The dashed line shows the location of the interface. Everything inside the box belongs to the single particle model, and everything outside the box belongs to the comprehensive code framework. Under this proposal, AFR would

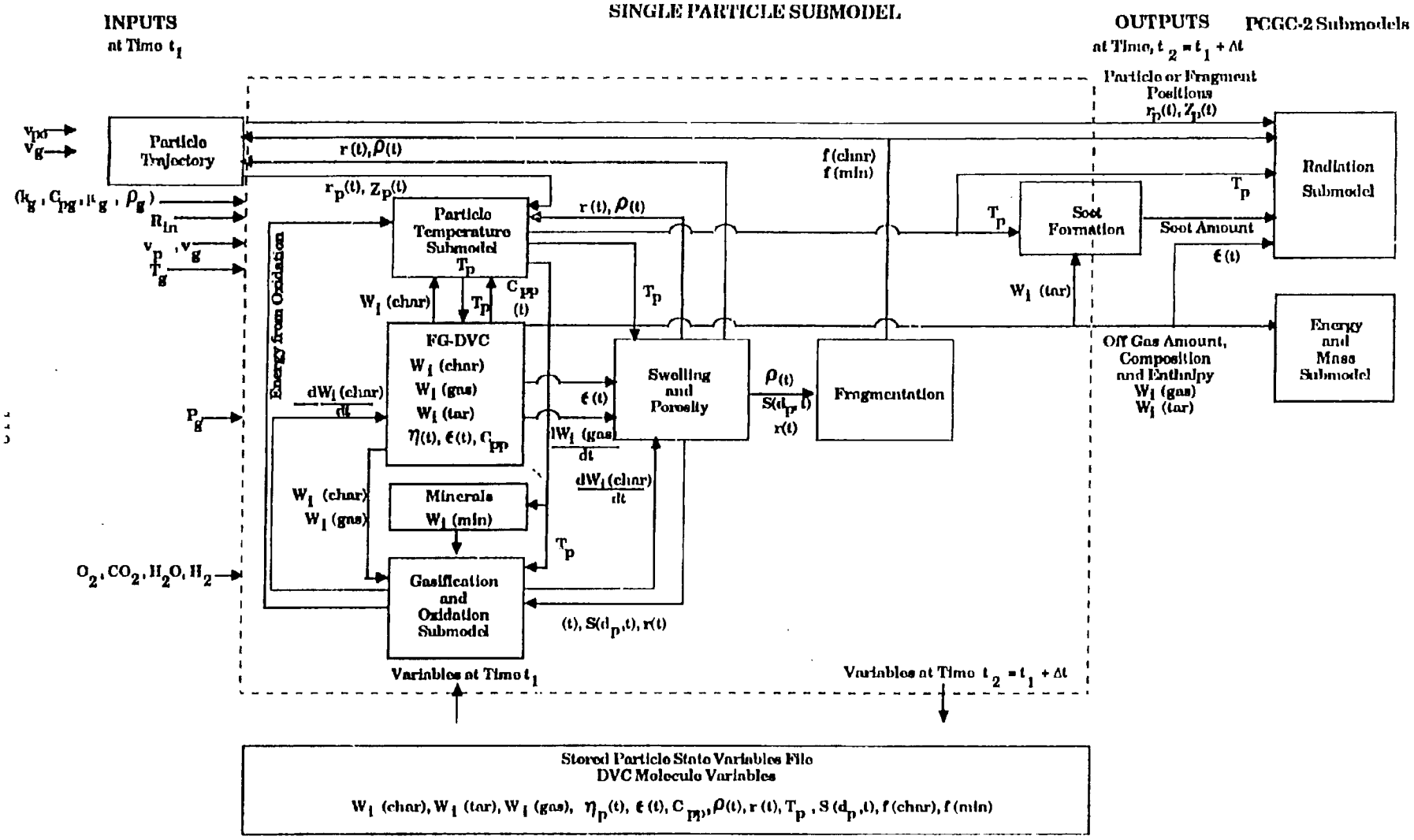


Figure III.A-2. Proposed Interface Between Single Particle Submodel and the Comprehensive Code.

take primary responsibility for the single particle model, and BYU would take primary responsibility for the comprehensive code framework. The proposed interface consists of inputs of particle and bulk gas properties at time t and outputs of particle, evolved gas, tar, and char properties at time $t+\Delta t$. Particle properties to be supplied would include composition, temperature, and diameter. The single particle model would operate with a particle state variable file storing the current DVC molecule variables from one call of the model to the next. The single particle model would solve the particle mass and energy balance equations for the entire reaction process, including devolatilization and heterogeneous reaction, but the particle trajectory calculations would be performed by the comprehensive code framework. Thus the single particle model would not require particle position in the reactor to perform its function. The single particle model would also determine particle diameter and extent of fragmentation, using swelling and porosity calculations. The proper location of soot formation with respect to the interface is not yet clear. If soot formation is to be treated as a particle phenomenon, it belongs inside the interface. If treated as a gas-phase phenomenon, it belongs in the comprehensive code framework.

The single particle model, as depicted in Figure III.A-2, would effectively replace the current PSOLVE and COAL2 subroutines shown in Figure III.A-1. The COAL1 and PSICT subroutines would remain essentially unaltered, except that the call to COAL1 would be made from PSICT rather than from PSOLVE. Also, PSICT would have to be altered to calculate the mass source terms to the gas phase based on parameters used in the FG/DVC model. PSICT currently treats raw coal and char as two separate components of the particle, with coal reacting to form char. However, the FG/DVC model treats the coal and char as a single component whose properties change gradually as the particle devolatilizes.

The advantage of incorporating the FG/DVC model under the SSPV approach is that it allows for the increased generality of the FG/DVC model, including varying offgas enthalpy, without significant complication in the comprehensive code framework. However, the approach is limited in that all elements in the coal must be assumed to evolve at the same relative rate. Allowing for

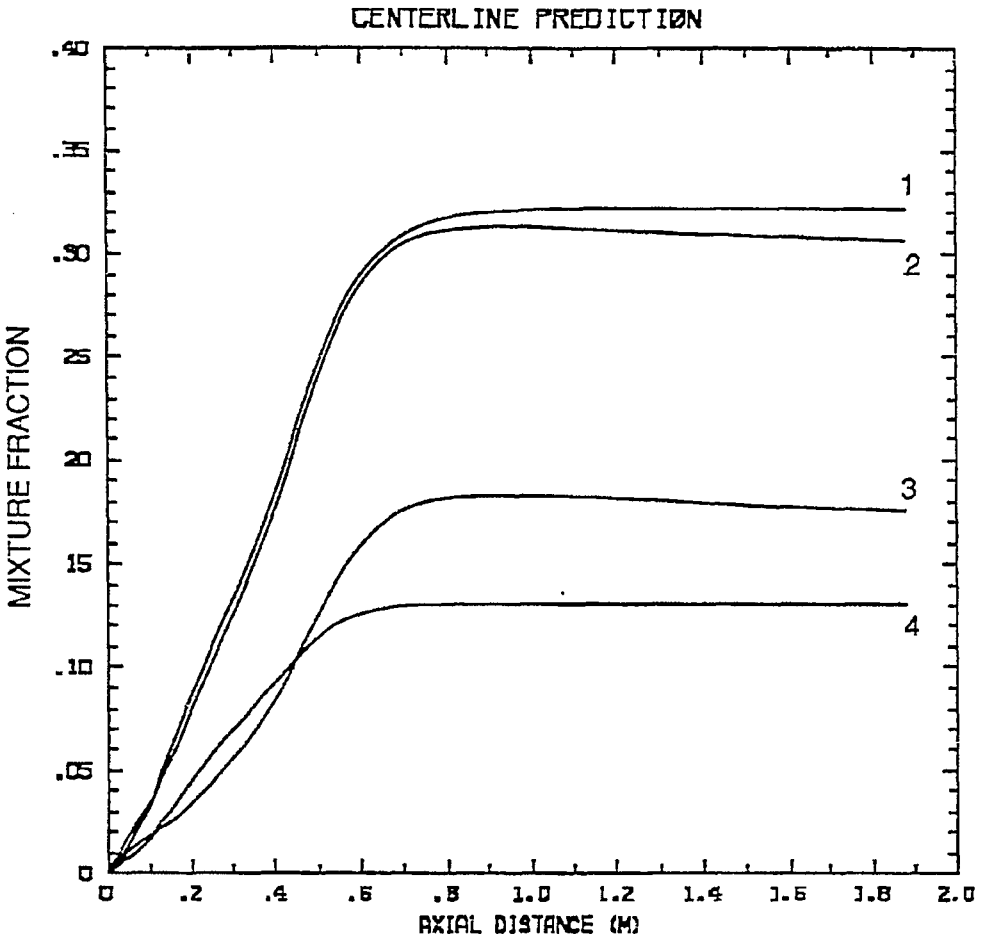
different relative rates requires additional progress variables or a new approach for modeling chemistry/turbulence interactions.

Multiple Solids Progress Variable (MSPV) Method -- This method allows the coal offgas elemental composition to vary with extent of burnout. Hence, hydrogen can be allowed to evolve more rapidly than carbon, for example, and nitrogen can be allowed to evolve more slowly. The evolution rate of nitrogen is particularly important, because of its propensity to form nitrogen oxide in the presence of oxygen. Nitrogen evolved in fuel-rich regions of the reactor forms molecular nitrogen rather than nitrogen oxides. Hence, accurate prediction of the nitrogen evolution rate from the coal is prerequisite to accurate prediction of nitrogen oxide level in the product gas.

In the MSPV method, each element may be tracked independently, or elements that evolve at similar rates may be lumped and tracked as a group. An additional progress variable is required for each additional independent element or group. The interaction of chemistry and turbulence is accounted for by integrating the instantaneous properties of the gas over the joint probability density function of all mixture fractions to calculate the time-mean properties.

The MSPV method is being tested in a limited fashion by using the two progress variables in the code to track coal offgas. Code modifications to permit this calculation were described in the first annual report (Solomon et al., 1987). During the last quarter, several bugs in the code have been identified and corrected, and a converged solution has been obtained where one progress variable was used to track coal volatiles and the second was used to track heterogeneous oxidation offgas. The single-rate model of Solomon et al. (1986) was used for devolatilization. The char oxidation offgas was assumed to be pure carbon. Using the two progress variables in the current code to both track coal offgas is limited to cases where the primary and secondary gas streams are identical in both composition and temperature.

Centerline mixture fractions for both one and two progress variables tracking coal offgas are shown in Figure III.A-3. The top curve represents the coal gas mixture fraction for the case where a single progress variable



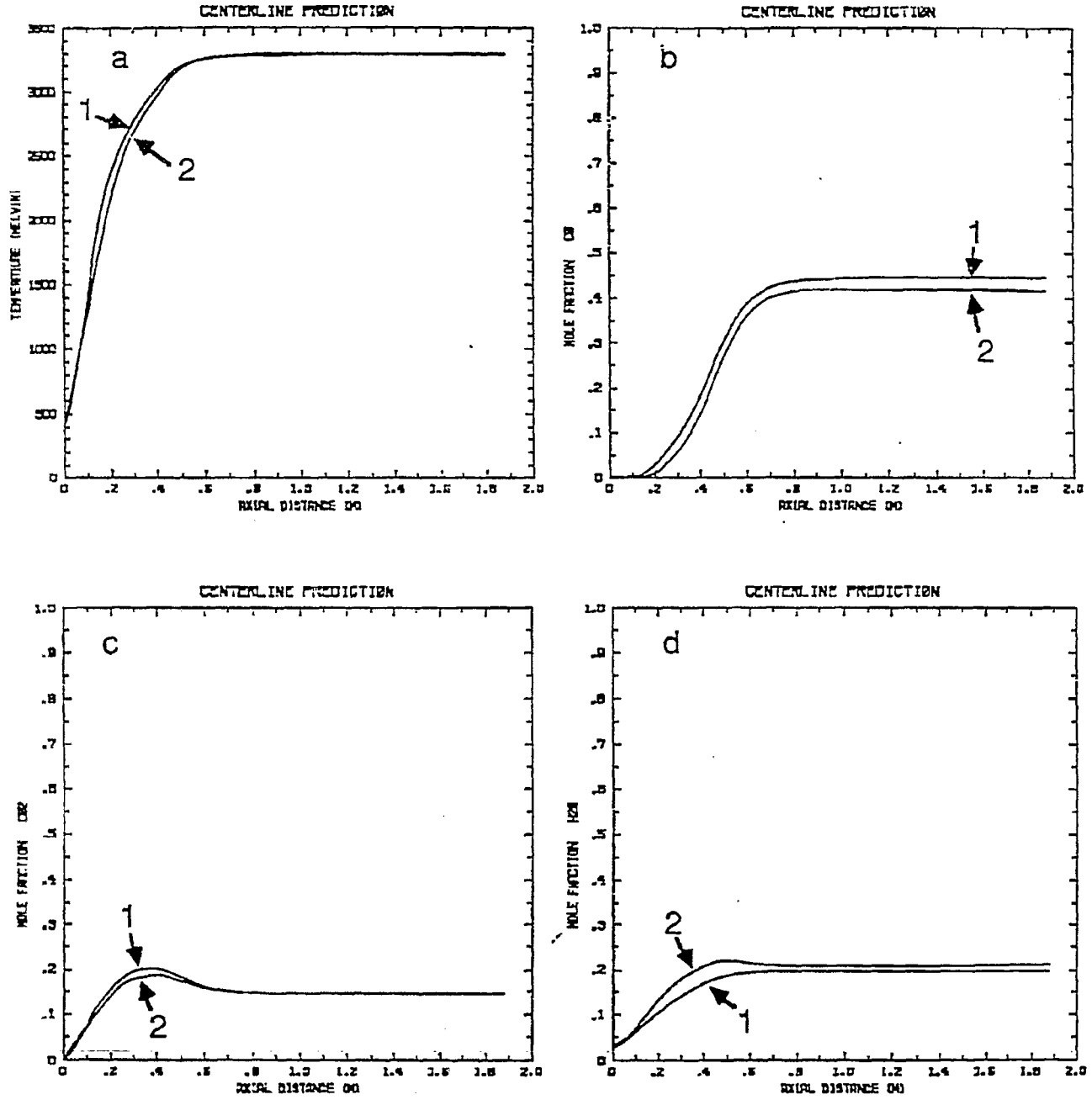
Curve	Description
1	Coal gas mixture fraction for the case where only a single coal gas mixture fraction was used.
2	Total coal gas mixture fraction for the case where two coal gas mixture fractions were used. Obtained by adding Curves 3 and 4.
3	Char oxidation offgas mixture fraction for the case where two coal gas mixture fractions were used. Based on the total gas mixture.
4	Coal volatiles mixture fraction for the case where two coal gas mixture fractions were used. Based on the total gas mixture.

Figure III.A-3. Coal gas mixture fractions for cases of one and two progress variables tracking coal offgas.

was used to track coal offgas. The three lower curves represent the total, char oxidation, and volatiles offgas mixture fractions, respectively, for the case where two mixture fractions were used to track coal offgas. Curves 3 and 4 are both based on the total gas mixture. Hence, Curve 2 is the sum of Curves 3 and 4. Comparing curves 1 and 2, the use of two progress variables to track coal volatiles and char oxidation offgas makes a small difference in the total coal offgas mixture fraction at the centerline. Comparing Curves 3 and 4, the oxidation mixture fraction exceeds the volatiles mixture fraction at an axial distance less than ~0.1 m. This is due to heterogeneous oxidation of the raw coal. However, the volatiles mixture fraction increases rapidly as the particle heats up and begins to devolatilize, overtaking the char offgas mixture fraction at a distance of ~0.1 m. After devolatilization is complete, the char oxidation mixture fraction continues to increase and overtakes the volatiles mixture fraction at a distance of ~0.45 m. Both Curves 2 and 3 decrease slightly with increasing axial distance after burnout is essentially complete (at ~0.8 m). The reason for this decrease is not yet clear. It may be due to radial mixing. No such decrease was noted in the case of a single progress variable (Curve 1), however.

The effect of tracking volatiles and oxidation offgas separately on centerline temperature and concentration of several gas species is shown in Figure III.A-4. Differences in centerline temperature of up to ~300 degrees K are noted, with the single-progress-variable case being the higher, as shown in Figure III.A-4a. Centerline mole fractions of carbon monoxide, carbon dioxide, and water (steam) are also affected to varying extents, as shown in Figures III.A-4b, c, and d.

Review of Coal Reaction Submodels -- A coal devolatilization model being developed at The University of Utah (Grant et al., 1988) and based on percolation theory is currently being reviewed. The Grant model does not yet treat many of the important phenomena of the devolatilization process, such as transport and pressure effects and crosslinking, but it does offer some unique features that may be useful. One is the incorporation of NMR data to help define coal structure, and another is the use of percolation theory to account for lattice statistics analytically. NMR data may be useful as a replacement or supplement to the solvent-swelling data that are currently used by the DVC



1 = Single coal gas mixture fraction
2 = Two coal gas mixture fractions

Figure III.A-4. Effect of tracking coal volatiles and char oxidation offgas separately on gas temperature and concentration of several gas species at the reactor centerline.

model, and percolation theory provides closed-form, analytical solutions for lattice statistics that may provide a useful alternative for the Monte Carlo approach currently used in the DVC model. The closed-form solutions provided by percolation theory, however, are not as general as the Monte Carlo simulations.

Component 2 -- Incorporating Improved Numerical Solution Methods

The purpose of this component of the subtask is to consider incorporating improved numerical solution techniques that are being developed under separate funding (Smith and Smoot, 1987; Hedman et al., 1987), in this laboratory. No new or improved techniques are being specifically developed under this study. The new methods include improved numerical solvers, distributive relaxation, multigridding, and techniques to couple equations. They hold significant potential for increasing the robustness and speed of the comprehensive code. It is anticipated that these techniques will not be available for incorporation until Phase II of the study; hence, no work was performed on this subtask component during the past quarter.

Component 3 -- Incorporating SO_x-NO_x Submodel

The aim of this subtask component is to incorporate the SO_x-NO_x submodel being developed under Subtask 2.g into the comprehensive code, and to extend the comprehensive code to include sorbent injection and sorbent chemistry. Work was initiated during the past year on extending the existing pollutant model to include formation of thermal NO_x and programming this extension into PCGC-2. This effort is described under Subtask 2.g in this report. The modification to include sorbent injection and chemistry will be based on work being conducted under separate funding at the University of Utah.

Component 4 -- Implementing the Code on Computers

The aim of this component of the subtask is to implement the comprehensive code on several computers, including a workstation. This implementation will require, at a minimum, standardizing the source code so

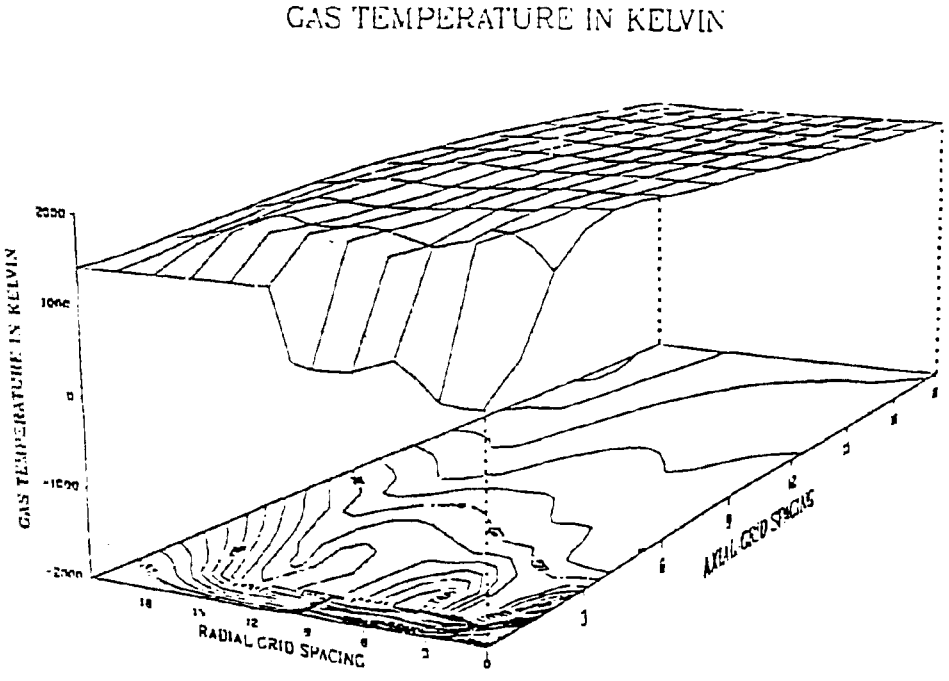
that it will run on a variety of computers. A user-friendly graphics interface is also desirable.

During the last quarter, a new research version of PCGC-2 (Baxter, 1987) was installed on the Sun-3/260 workstation at AFR, and a graphaical demonstration of PCGC-2 output was prepared and presented at the annual contract review meeting in November. The annual report described the selection of the National Center for Atmospheric Research (NCAR) software for demonstration, however problems were encountered implementing the NCAR software on the Sun, primarily due to a lack of adequate documentation. Therefore, the demonstration was prepared using the DISPLA software.

The new research version of PCGC-2 contains a statistical particle dispersion model and a generalized devolatilization model framework consistent with the Functional Group (FG) model. Simulation results for two fuel-lean cases were presented at the annual meeting, one using the FG model and the other using the two-step model with kinetics of Kobayashi et al. (1977). Reactor temperature maps produced by DISPLA for the two cases are shown in Figure III.A-5. The temperature fields for the two cases are very similar, however differences can be noted, such as the temperature trough in the corner of the reactor at the front wall in the case using the two-step model. No such trough was predicted for the case using the FG model. The flame structure can also be seen to be slightly different in the region near the secondary inlet.

Subsequent to the graphics demonstration using DISPLA at the annual meeting, UNIRAS was selected for future use with PCGC-2. This selection was based on an extensive evaluation of commercially available graphics software, including DISPLA. The evaluation was conducted at BYU by the Combustion Computations. Laboratory of the Advanced Combustion Engineering Research Center. UNIRAS was selected because of its user-friendliness, high level of support, and color capability.

a)



b)

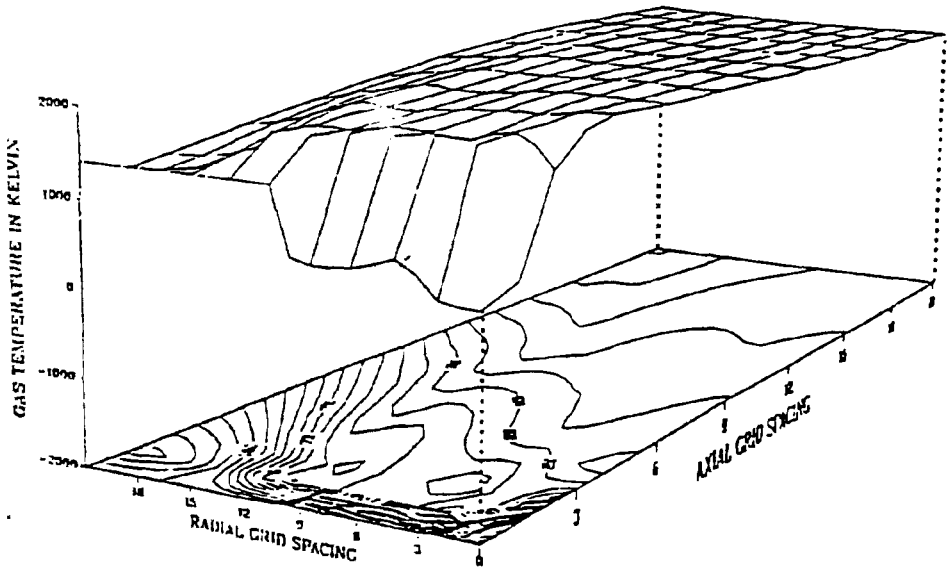


Figure III.A-5 Reactor temperature maps for a fuel-lean case using a) the FG model and b) the two-step model with kinetics of Kobayashi et al. (1977).

Component 5 -- Code Evaluation

The goal of this subtask component is to perform a statistical sensitivity analysis of input parameters to the improved code with advanced submodels and numerical methods incorporated under other components of the subtask. An existing databook will be used as a basis for the evaluation. No work was accomplished specifically under this subtask component during the past quarter, although the databook was revised and updated under separate funding (Christensen et al., 1987).

Plans

Work will continue during the next quarter on the three methods of integrating the FG/DVC model into PCGC-2. The particle model in the new Baxter version of PCGC-2 will be reviewed and compared with the 1987 version. An appropriate version will be chosen for integration and an interface for the single-particle model identified. Then, the FG/DVC model will be integrated, allowing coal offgas enthalpy to vary.

Calculations with two progress variables tracking coal offgas will continue to investigate the effects of tracking various elements separately and ignoring chemistry/turbulence interactions. Consideration will also be given to developing the theory of the Statistical Gas Dispersion (SGD) Model, eliminating the need for time-consuming calculations that convolve gas properties over the mixture fractions.

The review of the Grant devolatilization model utilizing NMR data and percolation theory will continue. Progress on the development of improved numerical solution methods in this laboratory will continue to be monitored for subsequent incorporation into the code being developed for this study.

Evaluation of the extension for thermal NO_x formation into PCGC-2 will be completed. Incorporation of the sorbent-SO_x submodel developed at the University of Utah will be initiated.

III.B. SUBTASK 3.B. - COMPREHENSIVE FIXED-BED MODELING REVIEW, DEVELOPMENT, EVALUATION, AND IMPLEMENTATION

Senior Investigators - Sung-Chul Yi, B. Scott Brewster, and L. Douglas Smoot

Brigham Young University
Provo, Utah 84602
(801) 378-2076, (801) 378-6240, and (801) 378-4326

Objectives

The objectives of this subtask are: 1) to provide a framework for an improved fixed-bed model that can incorporate coal chemistry submodels, improved boundary conditions, and pollutant formation processes; and 2) to provide a basis for evaluating the model.

Accomplishments

Phase I of this subtask has two components: 1) A literature review and evaluation of existing fixed-bed coal gasification models and experimental data, and 2) development of a proposed advanced model. During the last quarter, the proposed features of the advanced model were reviewed by external consultants. Dr. Charles Thorsness (Lawrence Livermore National Laboratory) and Professor Babu Joseph (Washington University) participated in the review. Based on their written comments, an extensive rationale and plan for developing the model have been formulated. A written development plan for the fixed-bed model was presented to AFR and METC personnel at the First Annual Contract Review Meeting. Mr. Justin Beeson was asked to respond with METC's approval and/or recommended changes.

An offer was accepted by a visiting scholar to come to BYU for at least a year and assist with the fixed-bed model development. Dr. Predrag Radulovic is currently Chief Research Engineer at the Institute of Thermal Engineering and Energy Research in Beograd, Yugoslavia. He will begin work in January and be responsible for the physical behavior of fixed beds and data for model validation.

Detailed accomplishments for each subtask component are described below.

Component 1 -- Literature Review and Evaluation

This subtask component is aimed at 1) reviewing existing models for fixed-bed coal gasification to determine elements that might be useful for developing the advanced model, and 2) locating experimental data that can be used for model validation.

Review of Existing Models -- A detailed review of existing models was accomplished previously and described in the first annual report. Based on the review, a recommendation was made that an advanced model be developed.

Evaluation of existing models continued during the last quarter. Predictions obtained from the Washington University 2-D model were compared with experimental values, and a sensitivity analysis was performed. The model was run with Illinois bituminous coal (low reactivity) and Wyoming subbituminous coal (high reactivity). The ultimate and proximate analyses of these coals were obtained from Cho (1980). Oxygen-blown, dry-ash operation of a Lurgi gasifier was modeled. Specific operating conditions for each case were obtained from Yoon et al. (1978). Comparisons of the predicted product compositions with experimental data for the Washington University (WU) and University of Delaware (UD) 1-D and 2-D codes are given in Figures III.B-1 and III.B-2.

For the Illinois coal, the WU 1-D model has been found to predict too much hydrogen, too little carbon dioxide, and too little methane in comparison with plant data (Yoon et al., 1978). The results of the WU 2-D model give some improvement. Overall product gas distribution predicted by WU 2-D model is best among the four sets of model predictions.

There are no experimental data available for the Wyoming coal case. Therefore only model predictions are shown in Figure III.B-2. The WU 2-D model predicts less hydrogen and more methane than the other models. Also, the predicted carbon dioxide is higher than for the other three models.

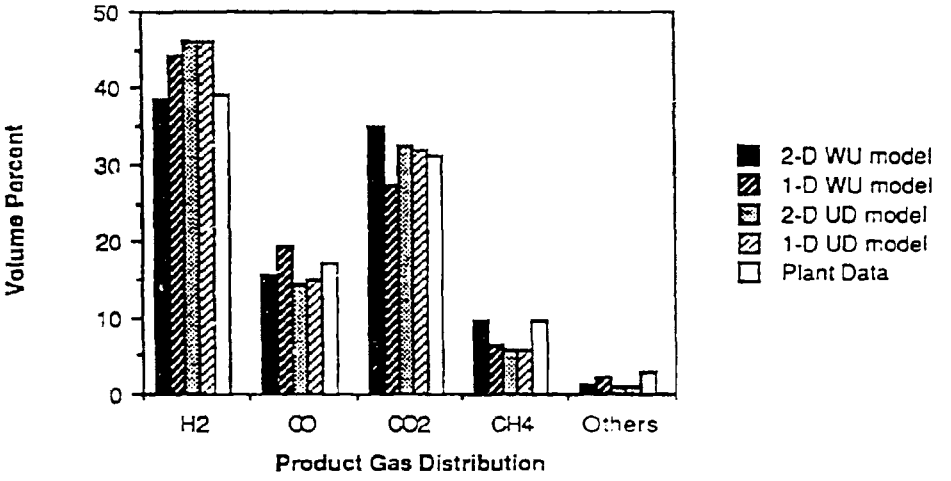


Figure III.B-1. Comparison of product composition predicted by various codes with plant data for Illinois coal.

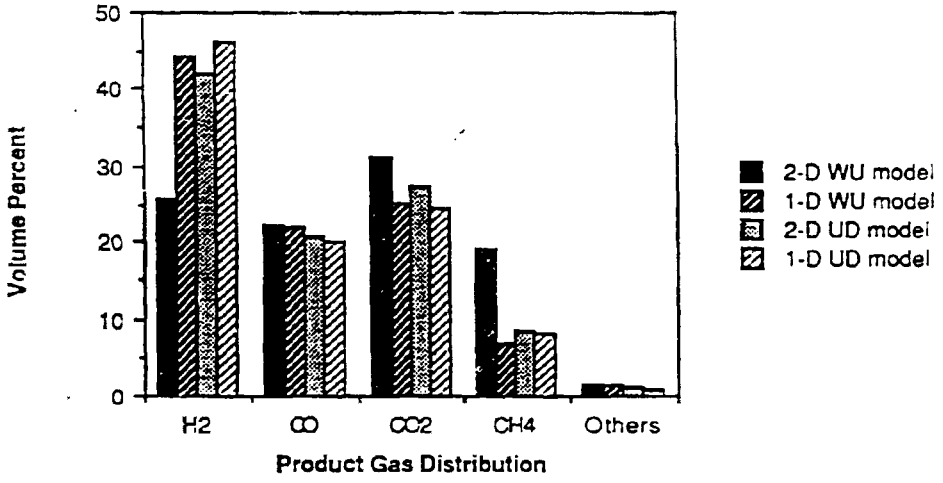


Figure III.B-2. Comparison of product composition predicted by various codes for Wyoming coal.

Before conclusions are made on the WU 2-D model, more simulations of different ranks of coal and different gasifier types should be made.

The sensitivity analysis was performed on gas inlet temperature, heat transfer coefficient, and wall temperature. As input to the WU 2-D model, the inlet gas temperature must be specified. The inlet temperature was varied from the base case of 644 K to 544 K and 744 K. The effect of gas inlet temperature on product gas composition is shown in Figure III.B-3. As inlet temperature increases, hydrogen and carbon monoxide increase while methane and carbon dioxide decrease. Methane and hydrogen changed less than 1% over the 200 K temperature range. However, carbon dioxide and carbon monoxide changed significantly. The WU 2-D model assumes that the molar ratio of carbon monoxide/carbon dioxide in the combustion reaction follows an Arrhenius temperature relation (Rossberg, 1956). The relation used in the WU 2-D model is:

$$[CO]/[CO_2] = k_0 \exp(-E/RT_g) \qquad \text{(III.B-1)}$$

where $[CO]/[CO_2]$ is the molar ratio of carbon monoxide to carbon dioxide and T_g is the gas temperature. Thus, changes in T_g significantly impact the CO/CO_2 ratio.

Increasing the wall temperature from the 496 K to 796 K had virtually no effect on the maximum temperature. Figure III.B-4 illustrates that increasing the wall temperature by 200 K did affect the outlet gas temperature, increasing it by 80 K.

The base case was run with a heat transfer coefficient of 340 kJ/m^2-hr-K. Decreasing the value of the coefficient to 140 kJ/m^2-hr-K reduced the amount of heat loss to the surroundings and increased the outlet temperature by 15 K. However, the outlet temperature decreased by 20 K when the heat transfer coefficient was raised to 540 kJ/m^2-hr-K, as shown in Figure III.B-5. Maximum temperature was fairly insensitive to radiation in heat transfer coefficient.

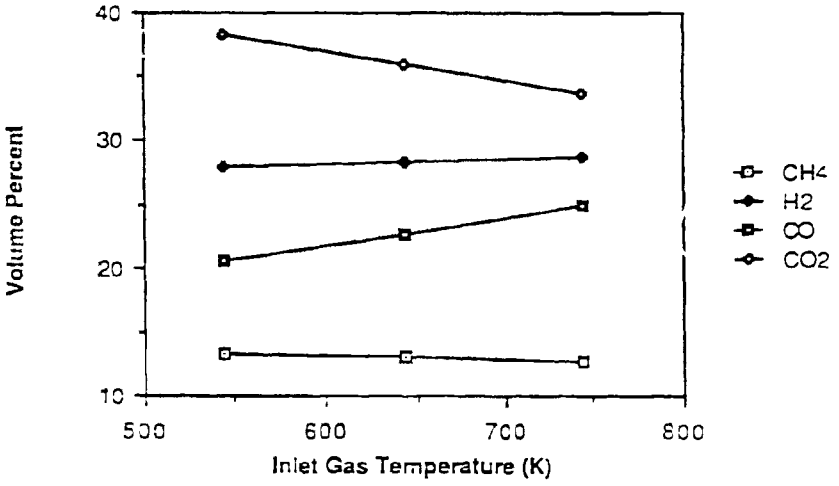


Figure III.B-3. Effect of gas inlet temperature on gas composition

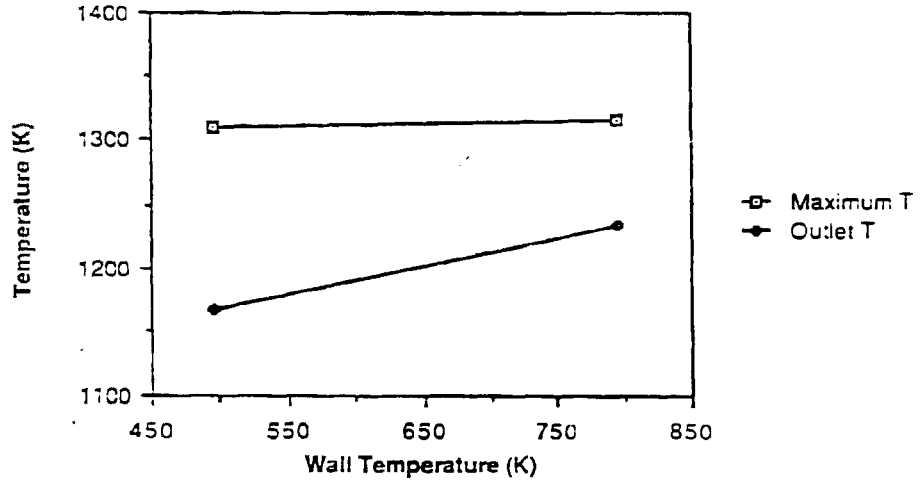


Figure III.B-4. Effect of wall temperature on gasifier temperature

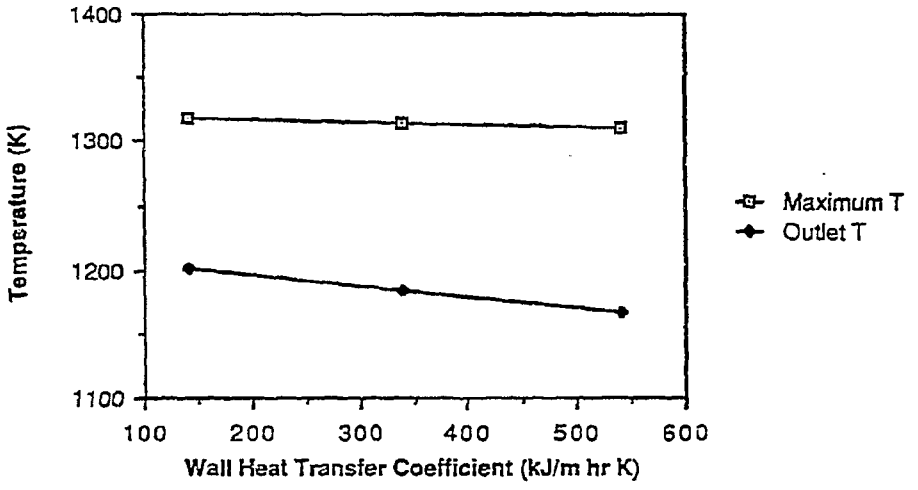


Figure III.B-5. Effect of heat transfer coefficient on gasifier temperature.

Component 2 - Detailed Plan for the Fixed-Bed Model

The purpose of this component of the subtask is to develop an advanced fixed-bed model. Based on the review of existing models, it was concluded that the model should not be based on an existing model, but that a new model should be developed using appropriate elements of existing models. A research plan for developing the advanced code was formulated under Subtask 1.b and presented to METC and AFR at the First Annual Contract Review Meeting, which was held in Hartford, Connecticut, in November 1987.

A detailed explanation of the advanced model features and a development schedule are contained in the research plan which was submitted to METC. Generalizing coal reaction processes, extending the gas phase to consider more species, treatment of pollutants and radiation, and improved treatment of solids and gaseous flow processes are the principal areas of focus. The rationale for developing an advanced model includes the following:

- The past level of effort in fixed-bed modeling has been quite modest.
- There is currently no other known ongoing investigation in fixed-bed modeling.
- There is currently no generalized, robust, well-documented code for fixed-bed coal gasification available.
- There has been little evaluation and application of fixed-bed models.
- Fixed-bed technology is of considerable current interest in high pressure combined-cycle power generation, synthesis gas production, liquids production (mild gasification), fuel gas production, and combustion.
- The importance of fixed-bed technology was specifically noted in two COGARN (Penner, 1987) recommendations.