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

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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. During the last quarter, the FG/DVC submodel previously integrated into an older version of PCGC-2 by AFR was integrated into the current version, and the new integrated code was transferred to AFR. Testing and validation of the integrated code was initiated to identify areas for future improvement.

Integration of FG/DVC Submodel into PCGC-2 - The FG/DVC submodel previously integrated into an older version of PCGC-2 by AFR was incorporated in the current version of the comprehensive code. The submodel was incorporated as an option that can be selected by the user in the main input file. The option of using the former devolatilization submodel, a simple weight-loss model (single equation or competing reactions), was preserved. The FG/DVC submodel interfaces with PCGC-2 in three routines: the main program (PCGC-2), the particle-source-in-cell routine (PSICT) which solves the particle trajectories, and the coal reaction subroutine (COAL2). An information flow diagram is shown in Figure III.A-1 to illustrate these three areas of interface. The main program calls routines to read the coal datafile, FG kinetics datafile, and DVC datafile, and to initialize the FG pools. After solving the gas phase equations (steps 2, 3, and 4), the particle number density fields (step 5), and radiation field (step 6), the Lagrangian particle calculations are performed. As shown, each particle trajectory is initialized and solved in turn. This initialization includes that for the FG/DVC model calculations, including construction of model oligomers for the DVC model. The FG/DVC initialization routines are called in PSICT. The particle continuity equations are solved using a predictor-corrector method, where two calls are made to the coal reaction subroutine, COAL2, to calculate the coal reaction rate at each time step. Since the FG/DVC submodel updates model oligomers and functional group compositions for each call, only one call is allowed to FG/DVC per time step. This call is performed in COAL2 during the corrector step.

A description of the subroutines that comprise the FG/DVC submodel is given below:

AMNT . . . .Calculates the change in each additional group pool for a time step.

BNDLOC . . .Randomly locates a bond of a given type in the bonds array.

BONDR. . . .Returns the bond type for a specified bead and bond site in the bonds array.

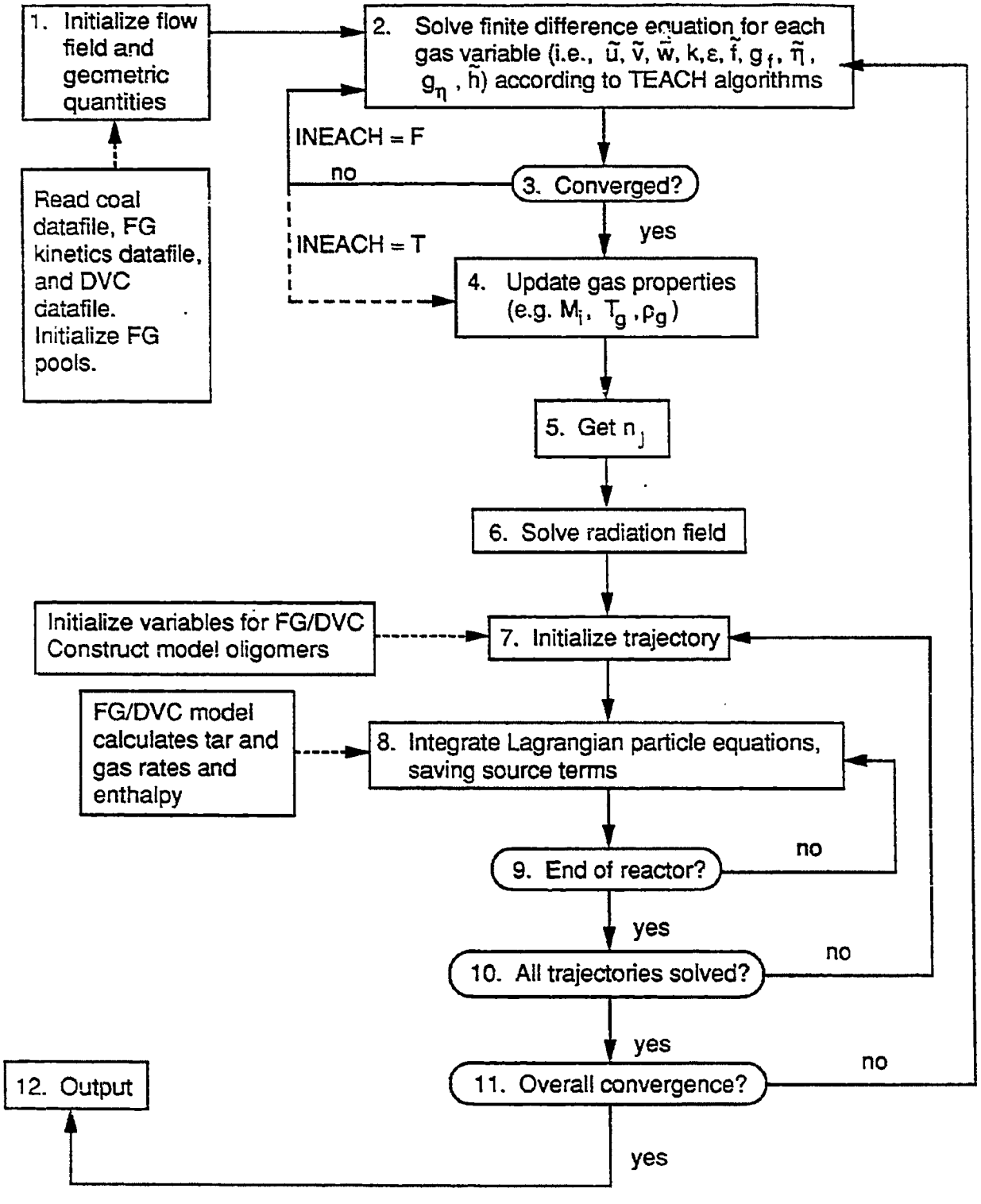


Figure III.A-1. Information flow diagram for PCGC-2 code with integrated FG/DVC submodel.

BONDW. . . .Writes to the bonds array for a specified bead and bond site, recording the bond type.

DATIN. . . .Reads the input data for the DVC model.

DECOD. . . .Reads data from input files.

DVC. . . . .Driver subroutine for DVC model. Randomly inserts crosslinks, eats away part of the oligomers to account for the evolved gas, and calls MONTE to perform Monte Carlo calculation.

FINFL1 . . .Called by COAL2 on corrector step of predictor-corrector algorithm in PCGC-2 particle model to add up amounts of various gas species and tar that have been produced thus far or that still remain in the char, calculate the molecular weight of the char, and get ready for the next iteration.

FINFL2 . . .Not used in the current implementation.

FKAYS. . . .Not used in the current implementation.

GETCDF . . .Reads the coal datafile.

GETKIN . . .Reads the kinetic datafile for the FG model.

HEAT . . . .Calculates the rate of released heat of pyrolysis, rate of volatiles enthalpy, rate of char evolution, and heat of combustion of char.

INIPOL . . .Called at the beginning of each particle trajectory calculation in PSICT to initialize variables for the DVC model.

INITYO . . .Called at the beginning of the main program, after the coal datafile is read, to initialize the mass fractions of the FG pools.

KBAR . . . .Calculates average reaction rates for each functional group.

MONTE. . . .Performs the Monte Carlo simulation of depolymerization, vaporization, and cross-linking.

NFCN . . . .Called by THER15 to calculate new distribution functions and the fractional change in each FG pool for a time step.

OLIGUP . . .Called by RXN1 to update the oligomer records when a bond is broken.

OLIMER . . .Adds or subtracts specified oligomers from their mass class.

OLIVAP . . .Called by MONTE to correct the mass concentrations of oligomers and the bonds concentrations after an oligomer is vaporized.

PER12. . . .Called by HEAT to calculate elemental composition of the char.

POLYBD . . .Called by INIPOL at the beginning of every particle trajectory to construct the model oligomers for the DVC model.

REINIT . . .Called by PSICT at the beginning of every particle trajectory to reinitialize variables for the FG/DVC submodel.

RXN1 . . . .Called by MONTE to randomly break two adjacent ethylene bridges, form methyl groups at one, and a hard bond at the other.

RXN2 . . . .Called by POLYBD initially to randomly insert specified number of crosslinks in the starting "molecule", and then later by DVC to randomly insert crosslinks that are formed as carbon dioxide and methane are released in the gas. There is also a call to RXN2 in MONTE to do the same thing, but it is inactive in the integrated version of the model.

SPEED. . . .A function similar to AMNT that calculates the change in the FG pools in THER15 to help determine whether or not the time step in THER15 is small enough.

TBLRTS . . .Called once by COAL2 and thereafter by THERI5 to calculate the rates for the functional group decomposition reactions and set up the parameters used by KBAR and NFCN to calculate the average rate constants and fractional change in functional group composition for a time step, respectively.

THERI5 . . .Driver routine for the combined FG/DVC model. Calls NFCN to determine the fractional change in each functional group composition for a time step, updates the functional group compositions, calls DVC to calculate the tar evolution, updates the tar functional group composition to be consistent with the DVC calculations, redistributes the source terms for aliphatic C and H, methane, and aromatic C, resulting from the bond-breaking, and calculates the tar evolution rate used in HEAT.

VAPSET . . .Called by MONTE to calculate constants used in VAPVEL to calculate vaporization rate.

VAPVEL . . .Called by MONTE to calculate the vaporization rate.

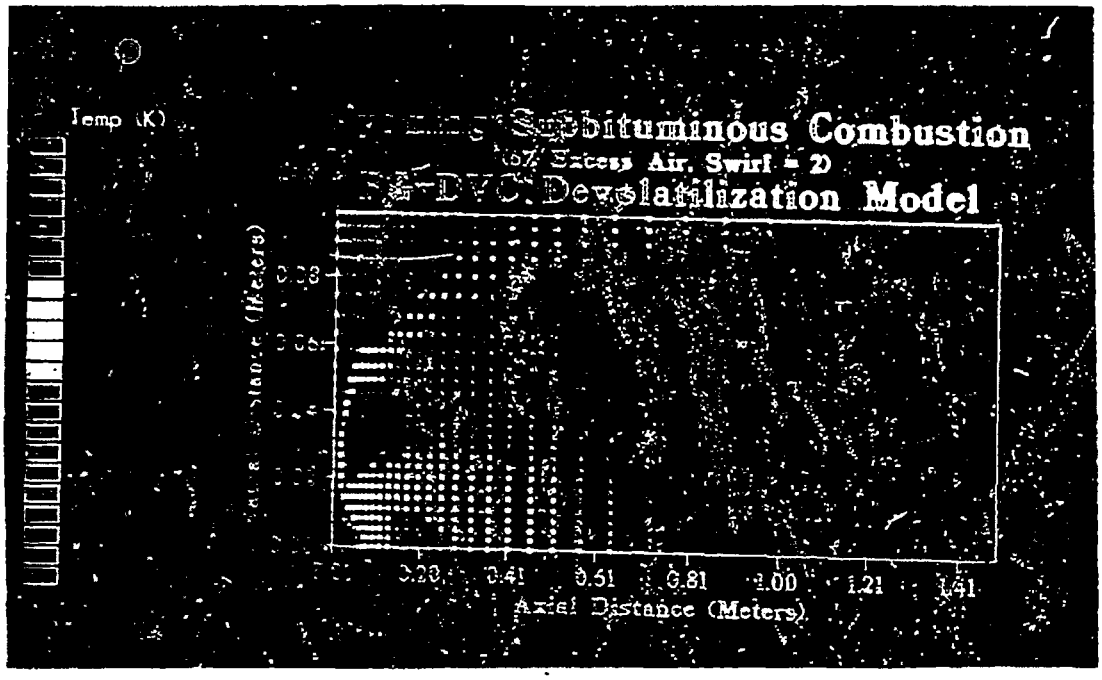
VISCOS . . .A function used by VAPVEL to calculate the viscosity of the metaplast as a function of temperature and molecular weight. (Not currently implemented in the integrated version.)

VPSETO . . .Called by INIPOL at the beginning of a particle trajectory calculation to calculate constants used in the tar evaporation model.

WRTOUT . . .Called by PSICT at the completion of devolatilization for each particle trajectory to write out results to an output file.

Evaluation of the FG/DVC Submodel - The FG/DVC submodel has been exercised for a pulverized coal and the results have been plotted with the UNIRAS software package on a Sun-3 workstation. Color graphics plots showing the temperature at each node point are shown in Figure III.A-2, for the FG/DVC submodel compared with results obtained with the two-step, competing-reaction devolatilization

a)



b)

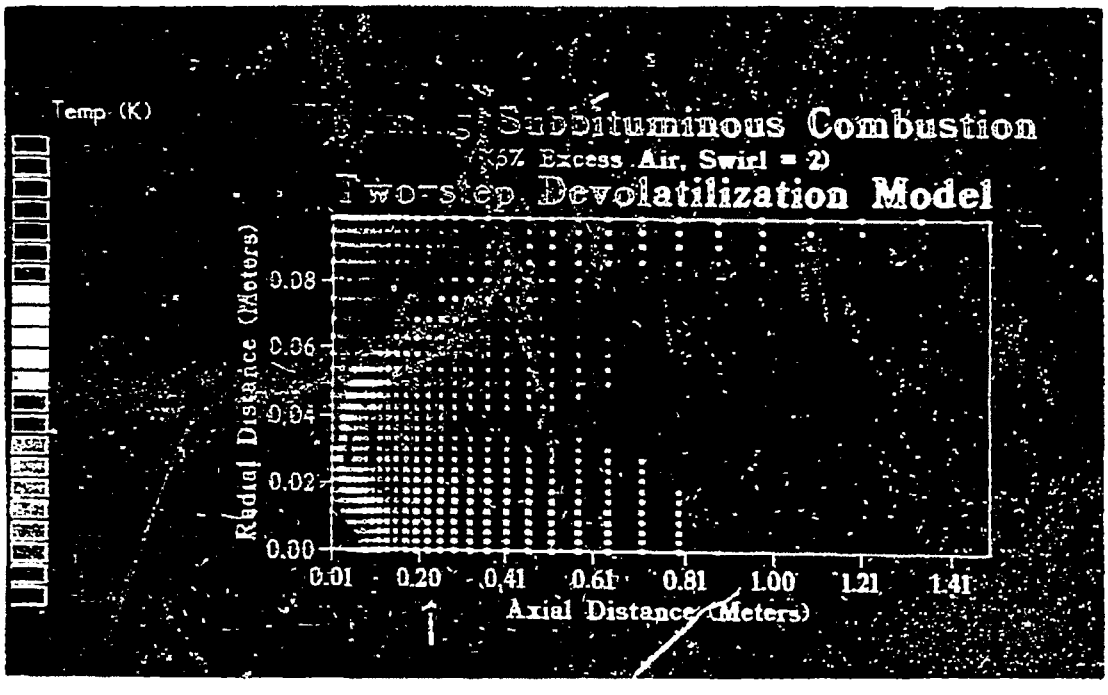


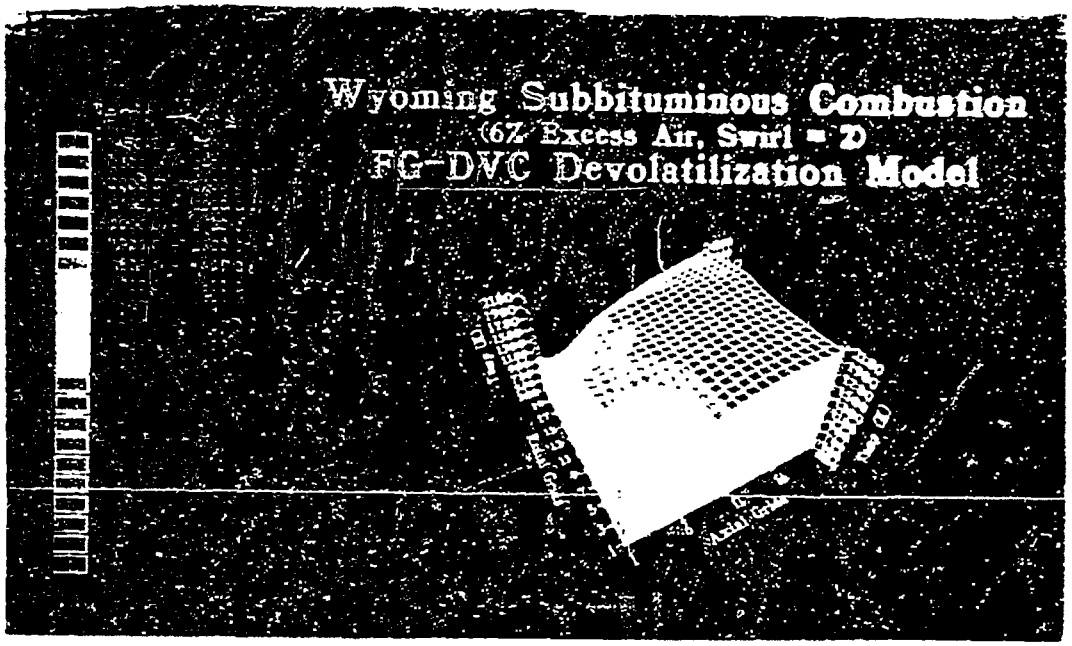
Figure III.A-2. Temperature plots for pulverized coal combustion case showing node locations.

submodel using the rate parameters of Kobayashi et al. (1977). The location of the dots in the figure corresponds to the location of the grid points. The onset of devolatilization occurs slightly earlier in the case of the FG/DVC submodel, as expected. Figure III.A-3a shows a surface plot where temperature is indicated both by the vertical distance and color. This type of plot is particularly useful for illustrating the correlation between two variables, where one variable can be shown by vertical distance and the other variable can be shown by color. The computational grid is superimposed on the surface in white. A third type of plot is shown in Figure III.A-3b. In this plot, coal gas mixture fraction (eta), inlet gas mixture fraction (f), and gas temperature are shown simultaneously by color on flat surfaces that are stacked vertically. This type of plot is also useful for illustrating correlations between variables.

A review of the FG/DVC submodel was initiated to identify areas of future improvement. One such area is the interface between the submodel and PCGC-2. The submodel uses properties of the bulk gas, such as viscosity, pressure, and diffusivity, that are currently supplied as input in the FG/DVC submodel input files or taken as fixed values in the submodel subroutines. The local bulk gas properties are determined by PCGC-2 in subroutine PSICT, and should be taken through the submodel interface rather than determined independently. It is also recommended that the units in the submodel all be converted to SI to be consistent with PCGC-2, and that the input be changed to directly correspond to the input variables described in the paper by Solomon et al. (1988). Variables that are not independent should be eliminated from the input.

Evaluation of Advanced Devolatilization Models - To evaluate the impact of advanced devolatilization models on advanced combustion codes, a generalized flame propagation model that describes pre-mixed, laminar gaseous and particle-laden flames is being investigated. The one-dimensional code, FLAME, is based on non-equilibrium chemistry. The incorporation of the FG/DVC model would allow evaluation that is independent of the fluid mechanics or turbulence model. Progress variables would not be required to track off-gas species. The code models the Combustion Laboratory's laminar premixed reactor which has many advanced diagnostic capabilities. Accurate gas temperatures and species concentrations will be measured with CARS. FLAME has been considered for

a.) Surface plot showing temperature both by color and by vertical height.



b.) Stacked plot showing gas mixture fraction (eta), inlet gas mixture fraction (f), and temperature.

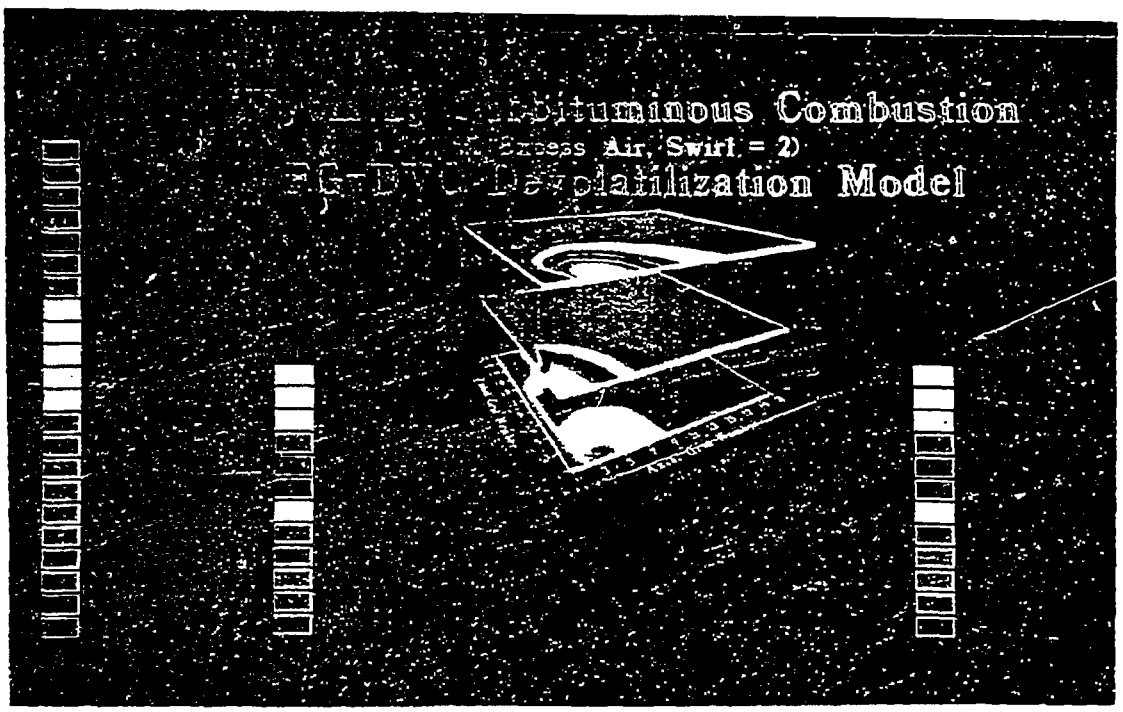


Figure III.A-3. Additional plots for pulverized coal combustion case with FG/DVC submodel.

incorporation of a NO_x model whic may enhance the evaluation of the NO_x model in the FG/DVC-integrated PCGC2. FL E has been installed on the Sun 3-260.

Statistical Gas Dispersion Mo - The statistical gas dispersion model is an advanced concept for treating e interaction of chemistry and turbulence without the complications of mixtu fractions and convolution integrals. The gas is treated in a Lagrangian fra of reference, with statistics. The method is based on that of Baxter (88) for particle dispersion, and will theoretically allow for variable offgas composition and enthalpy, with chemistry/turbulence interactions, but without nested integrations in order to calculate time-mean properties. This concept has potential for improving the integration of the FG/DVC submodel in PCGC-2. During the past quarter, initial plans were made for developing this concept. Application was made to Sandia National Laboratories in Livermore, California, for Mr. Michael Hobbs to visit the laboratory for a six-month period to work on the development of this method jointly with Mr. Larry Baxter.

Component 2 - Incorporating Improved Numerical Solution Methods

The purpose of this component of the subtask is to incorporate applicable improved numerical solution techniques that are being developed under separate funding by Smith and coworkers (Smith and Smoot, 1988) in this laboratory. During the last quarter, under separate funding, work continued on developing the multigrid method and incorporating it into a 3-D fluid mechanics code. This independent work will be documented later this year in the Annual Report of the Advanced Combustion Engineering Research Center and will be summarized in a future quarterly report.

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 continued on incorporating thermal NO in PCGC-2 as described under Subtask 2.g. The modification to include downstream injection of sorbent particles and their subsequent reactions with the gas phase is being based on independent work being

performed by Pershing and coworkers (Milne and Pershing, 1987) at The University of Utah, where a sorbent chemistry submodel for fuel-lean conditions is being developed and incorporated into PCGC-2. During the last quarter, work was initiated to prepare a version of PCGC-2 that has been tested on the Sun-4 workstation. This code will then be transferred to Utah where the sorbent particle submodel will be incorporated.

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. As described above, work was initiated on testing PCGC-2 on a Sun-4 workstation prior to transferring it to The University of Utah for incorporation of a sorbent particle submodel. Work was also initiated on the development of a color graphics interface for interpreting code output. Several color graphics figures have already been shown, illustrating predictions made with the FG/DVC submodel compared with the two-equation devolatilization model of Kobayashi et al. (1977). These figures were obtained with the UNIRAS graphics package on a Sun-3 computer. In addition to UNIRAS, a sophisticated color graphics package is being developed at BYU primarily for 3-D applications, but it will also be applicable to 2-D. A temperature plot of a combustion case with the two-step devolatilization model illustrated by the new BYU graphics package under development is shown in Figure III.A-4.

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 this 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.

Figure III.A-4. Color fringe plot for temperature illustrating color graphics capability being developed at BYU under separate funding.

Plans

During the next quarter, work will continue on validating and improving the integration of the FG/DVC submodel in PCGC-2. The code will be made functional to run on the Sun workstations as well as the Convex mini-supercomputer. The effects of variation in coal offgas enthalpy and composition will be investigated with two solids progress variables. Char oxidation offgas and volatiles will be tracked separately. Work will continue on obtaining data for code validation. The arrangement to develop the statistical gas dispersion model based on a Lagrangian description of the gas will be pursued with Sandia National Laboratory. A laminar version of PCGC-2 will be developed to assist AFR with laboratory data evaluation. Work will continue on the development of a color graphics interface for PCGC-2 with the integrated FG/DVC submodel on the Sun workstation. Independently funded work on the multigrid numerical method will continue to be monitored. A version of PCGC-2 functional on the Sun workstation will be sent to The University of Utah for incorporation of the sorbent particle submodel.

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

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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. Progress during the last quarter is described below for each of these components. During the last quarter, the computer code development for the improved, fixed-bed gasifier model was continued. A 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.

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 used as a starting point for developing the advanced model, 2) determining appropriate correlations and submodels for physical properties of fixed beds, and 3) locating experimental data that can be used for model validation. The review of existing fixed-bed models was described in the First Annual Report (Solomon et al., 1987) and the 5th Quarterly Report (Solomon et al., 1987). The preliminary reviews of flow, mass and heat transfer in fixed-beds as well as

fixed-bed technology were presented in the 6th Quarterly Report (Solomon et al., 1988).

Collection of Data - The collection of fixed-bed reactor design and test data was continued. Particular attention was paid to mild gasification data. UCC Research Corporation's mild gasification program will probably provide some of the needed test data. The Eighth Annual Gasification and Gas Steam Clean-up Systems Contractors Review Meeting at Morgantown was attended. A number of presentations and posters were devoted to fixed-bed gasification and in particular to mild gasification. The Morgantown Energy Technology Center was visited, including a tour of the METC fixed-bed reactor.

Comprehensive Review - A comprehensive review of fixed-bed combustion and gasification was initiated. The review will include a summary of experimental observations, large particle reaction rates, models for fixed-bed combustion and gasification processes, features of fixed-bed reaction processes and related technology. This review is an extension of the fixed-bed modeling review conducted under this project last year and is independently funded.

Component 2 - Detailed Plan for Fixed-Bed Model

This subtask component has already been completed.

Component 3 - Development of the Framework for an Advanced Fixed-Bed Model

The purpose of this component of the subtask is to develop a code framework for an advanced, fixed-bed model. As a basis for this framework, an improved fixed-bed model is being developed. The improved fixed-bed model has many of the basic features of an advanced model, such as separate gas and solids temperatures, but is simplified in its treatment of chemistry and numerical solution methods.

Improved Fixed-Bed Model - The details of the improved fixed-bed model can be found in the 6th Quarterly Report (Solomon et al., 1988) and elsewhere (Yi et al., 1988). The computer code development for the improved fixed-bed gasifier model started on a VAX-8600 computer; the development was continued on a Sun-3

computer. A part of the computer code developed on the VAX-8600 computer was transferred and tested on the Sun-3 computer. Difficulties were encountered in solving the energy equation. Difficulties were also encountered in using the IMSL routine GEAR because of its automatic change in step size in integration. The IMSL routine MOLCH was selected for further work. As an improved estimate, a two-step devolatilization model was formulated and included in the improved fixed-bed model.

Most existing fixed-bed models follow the scheme shown in Fig. III.B-1. It is usually assumed that drying and devolatilization are instantaneous. The char combustion and gasification processes are then modeled with various rate expressions. The improved fixed-bed model assumes a segregated but finite devolatilization zone. The particle submodel for this zone will not include all the details of heat and mass transfer; it will be based on the two-step model of Kobayashi et al. (1977) and Ubhayakar et al. (1977). Later, in the advanced, fixed-bed model, inclusion of the FG/DVC model for devolatilization will be considered.

The numerical method for the improved fixed-bed model is based on orthogonal collocation for the radial direction and finite difference for time; thus the set of the PDE's can be rewritten as a larger set of ODE's in axial direction. This set of equations was numerically integrated using the IMSL routine IVPG based on a Gear's method (Gear, 1971). However, this method did not work; the split boundary conditions seemed to be the cause of the difficulties. To avoid this, two adjustments were made. First, the solid velocity was neglected; this is justified by the fact that this velocity is very small. Thus the inlet boundary condition for the solid phase was not required. Second, another IMSL routine, MOLCH, was tried; this routine is based on the method of lines. The routine was tested to check its applicability to the improved fixed-bed model. The MOLCH routine proved better than the IVPAG routine for the problem at hand; it will solve both the boundary value and the initial value problems. This routine will be used to complete the development of the improved fixed-bed model. Difficulties encountered in solving the energy equations were traced to minor mistakes in the formulation of the energy equations.

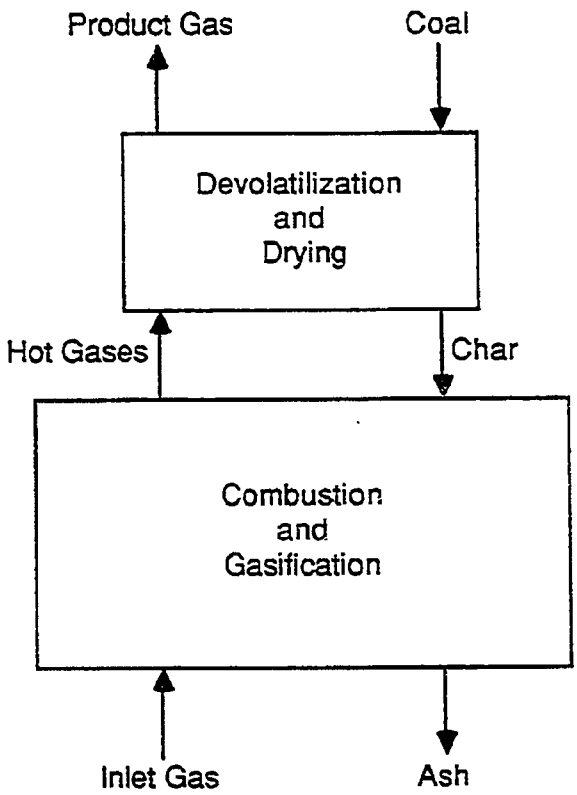

Figure III.B-1. Segregation of devolatilization and drying processes in fixed-bed gasifier modeling.

Advanced Fixed-Bed Model - The preliminary forms of the governing equations for the advanced, fixed-bed model were given in the Annual Report (Solomon et al., 1987) and in the Fixed-Bed Research Plan (Smoot et al., 1987). These equations are being rederived, based on the derivations of Crowe (1976) and Crowe and Smoot (1979), and extended to dense-phase systems. The equations are similar to those of the improved, fixed-bed model, but are being developed for a different set of assumptions. The equations and assumptions will be documented in a future report.

Plans

The improved fixed-bed computer code will be completed and debugged. The development of the advanced fixed-bed model will be continued. Particular attention will be paid to the flow of solids and the radiation heat transfer. The collection of the fixed-bed reactor design and test data as well as the work on the comprehensive review of fixed-bed combustion and gasification will also be continued.