FOSSIL FUEL CONVERSION--MEASUREMENT AND MODELING

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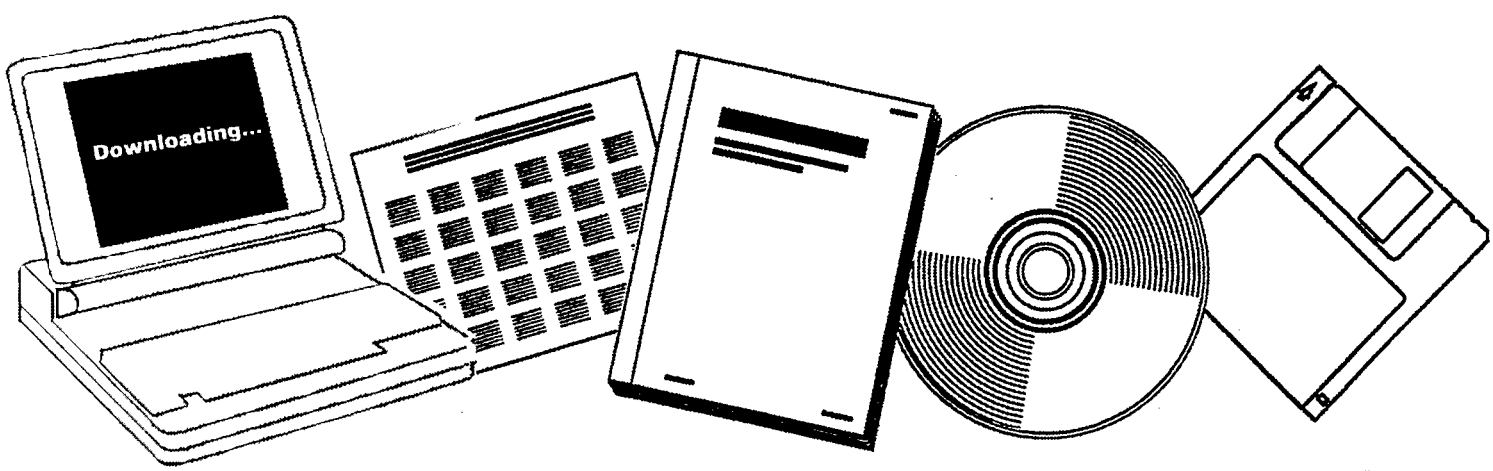

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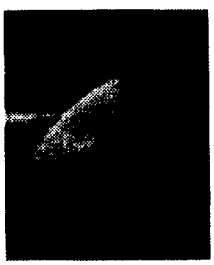

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Conference Location:

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DISCLAIMER

P17 Fossil Fuel Conversion - Measurement And Modeling

CONTRACT INFORMATION

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Period of Performance:	September 14, 1993 to September 13, 1996	

FY94 Program Schedule

		S	O	N	D	J	F	M	M	J	J	A
1.	Research Plan											
2.	Gasification Processes											
3.	Gas Phase Combustion											
4.	Fluidized-Bed Systems											
5.	Applications											

OBJECTIVES

The main objective of this program is to understand the chemical and physical mechanisms in coal conversion processes and incorporate this knowledge in computer-aided reactor engineering technology for the purposes of development, evaluation, design, scale up, simulation, control and feedstock evaluation in advanced coal conversion devices. To accomplish this objective, this program will: 1) provide critical data on the physical and chemical processes in fossil fuel gasifiers and combustors; 2) further develop a set of comprehensive codes; and 3) apply these codes to model various types of combustors and gasifiers (fixed-bed, transport reactor, and fluidized-bed for coal and gas turbines for natural gas).

BACKGROUND INFORMATION

To expand the utilization of coal, it is necessary to reduce the technical and economic

risks inherent in operating a feedstock which is highly variable and which sometimes exhibits unexpected and unwanted behavior. Reducing the risks can be achieved by establishing the technology to predict a coal's behavior in a process. This program is creating this predictive capability by merging technology developed at Advanced Fuel Research, Inc. (AFR) in predicting coal devolatilization behavior with technology developed at Brigham Young University (BYU) in comprehensive computer codes for modeling of entrained-bed and fixed-bed reactors and technology developed at the U.S. DOE-METC in comprehensive computer codes for fluidized-bed reactors. These advanced technologies will be further developed to provide: 1) a fixed-bed model capable of predicting combustion and gasification of large coal particles, 2) a transport reactor model, 3) a model for lean premixed combustion of natural gas, and 4) an improved fluidized-bed code with an advanced coal devolatilization chemistry submodel.

PROJECT DESCRIPTION

The program consists of five tasks: 1) Preparation of a Research Plan, 2) Modeling of Gasification Processes, 3) Modeling of Gas Phase Combustion, 4) Modeling of Fluidized Bed Systems, and 5) Applications. This paper describes the initial work performed during the first year of the contract.

RESULTS

Devolatilization of Large Coal Particles (AFR) - Large coal particles are typically used in fixed-bed combustion and gasification processes. The objective is to extend the Functional Group - Depolymerization, Vaporization, Crosslinking (FG-DVC) model that was developed for small coal particles to these centimeter size particles. Work is being carried out in three steps, 1) modeling heat transfer in a spherical coal particle, 2) development of a simplified coal devolatilization model based on FG-DVC, and 3) integration of the heat transfer model and the simplified coal devolatilization model.

Work started with the heat transfer model. Code development for the simplest situation with constant density, heat capacity and thermal conductivity and in the absence of pyrolysis was completed. The numerical solution is in good agreement with the analytical solution given by Jakob (1959). Future work will concentrate on integration of the FG-DVC pyrolysis model. Temperature dependent coal properties will be used in the code based on Merrick's thermal physical coal model (Merrick, 1983).

Reduced Version of Coal Devolatilization Model (AFR) - To model the devolatilization of large coal particles, it is necessary that the rate equations of coal devolatilization be appropriately expressed and solved along with the heat transfer equation. It is well known that coal devolatilization is a complicated process, and cannot easily be modeled with a single rate equation, especially when the elemental compositions of char, gas, and tar and the amounts of the individual gas species are desirable. The full FG-DVC model was implemented in the current version of the advanced fixed-bed code (FBED-1) in the previous contract, with the assumption that the particles are isothermal. Incorporating an intra-particle heat transfer model will require a substantial simplification of the FG-DVC model, in order to reduce the computational burden.

After a careful re-examination of the FG-DVC model, a number of model simplification options have been proposed. Option 1 is a single rate equation model which solves the total pyrolysis weight loss. Other time dependent variables, such as the yields of tar and gas species, are calculated by correlation with the weight loss. Option 2 has a single rate equation for total gas

yields and the DVC network submodel to predict the tar yield. Option 3 is Option 2 plus a submodel for nitrogen gas release, which cannot be evaluated accurately with the weight loss correlation due to its heating rate dependence. These options were constructed based on the following considerations:

1. Devolatilization of individual gas species occurs independently (except for NH_3, HCN) in the model.

2. Tar and gas evolution are coupled.

3. Tar evolution is influenced by pressure and heating rate. In order to model the pressure and heating rate dependence, the tar submodel in FG-DVC must be retained. However, this can be implemented in a pre-processing step so that the computational burden is not unduly increased.

It was therefore assumed that:

1. The total light gas evolution can be modeled using a single rate equation with a distributed activation energy accounting for multiple gas species contributions.

2. When the tar evolution amount is known from a pre-processing step, it can be treated as another "gas species" in addition to the light gas evolution, so the total volatile evolution can be modeled with a single rate equation.

3. Alternatively, the tar submodel can be solved along with the light gas species model, but it adds an additional computational burden.

4. The amounts of individual gas species can be recovered from correlation with the total volatile or total light gas amounts. This correlation is provided by the full FG-DVC model calculation which is run in a pre-processing step.

In summary, the light gas species can be modeled with a simplified model since these evolve independently, but the tar submodel cannot be easily simplified since the evolution depends on the process conditions (heating rate, pressure). With Option 1, the tar amount is estimated from the preprocessing step with FG-DVC based a nominal heating rate and bed pressure. Tar is then lumped into the total volatile equation and its contribution is modeled via its share in the activation energy distribution. In this case, the gas elemental composition is correlated with the total weight loss. The correlation is given by a standalone FG-DVC calculation and is an input to the one-equation model. The predictions of coal pyrolysis at a given heating rate by the one-equation model (Option 1) and by the full FG-DVC model are generally in good agreement.

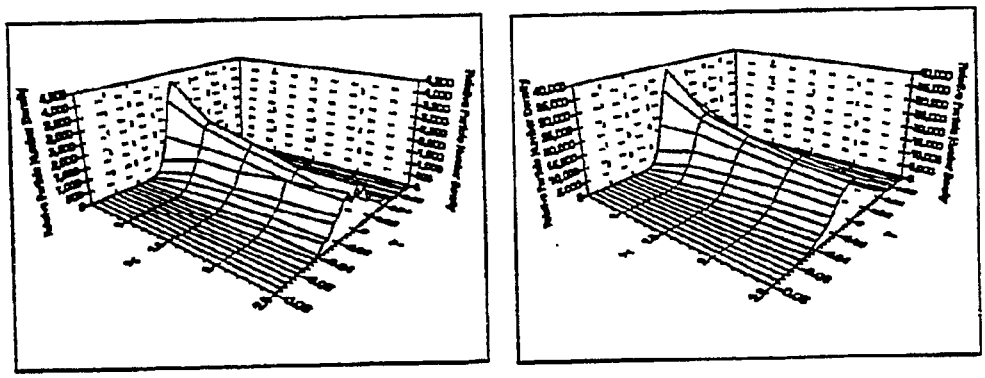
Transport Reactor Modeling (AFR) - To model transport reactors, we plan to start with the entrained-bed coal gasification code developed at BYU, PCGC-2. Since transport reactors have particle loadings which are much higher than entrained-beds, dispersion of particles and their interactions with the gas phase become important. A stochastic turbulence particle dispersion model is being developed and is being verified with available data. It is also recognized that there exist fluctuations of particle number density everywhere in the reactor. A model accounting for this effect has been outlined and will be implemented into our particle model. Literature reviews for data on chemical and physical processes in transport reactors for model validation are underway.

The work during the first year was focused on developing and verifying a standalone turbulence particle dispersion model. Once it is verified, it will be implemented in PCGC-2, replacing the existing empirical particle dispersion model. The code then will be executed to convergence for transport reactor conditions which feature moderate particle loadings. This model uses the Ornstein-Uhlenbeck stochastic process to simulate the turbulent component of the gas velocity. It was demonstrated that the turbulence generated has the exact values of k and the time scale given by the k-ϵ model. This is the advantage of our model in comparison with those of Gosman and Ioannides (1981), and Shuen et al. (1983). Their models used a direct Monte Carlo method to alter the local gas velocity and these numerical schemes will not necessarily give the correct values of k and ε. There are in-depth discussions on the numerical solution of stochastic equations which use the Ornstein-Uhlenbeck process by Kloeden and Platen (1982). Truncated Fourier series with independent Guassian random coefficients were also used to simulate the gas turbulence (for example, Wang and Stock (1992)), but computing a truncated Fourier series consumes large amounts of computer time. Our model is much more efficient.

In order to verify our model, predictions of particle dispersion need to be compared with existing data. One such comparison has been made with data in grid generated turbulence by Snyder and Lumley (1971). It was found that our model under-predicts the data. Shuen et al. (1983) indicated that their model was calibrated with Snyder and Lumley's data (1971), but the nature of this calibration is not clear. We agree with Shuen et al. (1983) that it may be due to the arbitrary estimation of the time and length scales of the turbulence. It is true that, although the Eulerian integration time and length scales do present the scales in which the turbulence develops, they may not be quantitative enough to be used in the turbulence particle dispersion model. Our model is clearly limited by the state-of-the-art in understanding gas turbulence phenomenon. Although this is a fast advancing field, the k-ε description is still the most commonly accepted model. Using other turbulence models is beyond the scope of this contract and we will continue to rely on the k-ε description. In order to fit Snyder and Lumley's data (1971), the time scale given needs to be increased by a factor of 2.7. The validation of this approach needs to be proved by comparing with more data.

Work was also carried out to calculate the Eulerian particle properties, such as the mean velocity and the particle number density, from the stochastic particle model. Obtaining statistically meaningful values requires a large number of particle trajectories. The distributions of particle number density are plotted in Figures 1a and 1b with 500 and 5,000 particle trajectory calculations, respectively. The calculation was based on Snyder and Lumley's fluid data mentioned above and particles were injected from a point source upstream. It is clear that, while the two calculations resulted in the same distributions, the distribution in Figure 1b is much smoother than that in Figure 1a. The insufficient number of trajectories which resulted in the noisy signals in Figure 1a, would cause severe convergence problem in the gas phase calculation, especially at high particle loadings. Since practical inlet pipes have finite diameters, model calculations have to inject particles from multiple positions throughout this diameter. Calculating 5,000 particle trajectories at each injection point imposes a heavy computational load on the code. We have to calculate only the number of trajectories, that is practically possible and look for ways to smooth the noisy distributions.

Fluidized-Bed Modeling (AFR) - The work on fluidized-bed modeling is directed toward developing a coal devolatilization submodel for an existing fluidized-bed code, MFIX. A simplified



a. b.

Figure 1. Relative Particle Number Density Calculated From the Stochastic Particle Dispersion Model. Gas Fluid Data are From Snyder and Lumley (1971). a) 500 Particle Trajectories and b) 5000 Particle Trajectories.

version of FG-DVC will be tailored toward the needs of fluidized bed models. Currently, the same options for model simplification and submodel specifications that are being developed for the Fixed-Bed Reactor code are also being considered for the fluidized code.

Oxidation Rates for Large Coal particles at High Pressures (BYU) - Fixed-bed combustion and gasification processes are typically done with large coal particles at high pressures but the previously available fixed-bed codes use kinetic rates derived from small particle experiments at atmospheric pressure. Thus, work began to measure kinetic rates for large coal particles at high pressures.

A unique experimental facility was constructed to obtain oxidation rates of large particles at high temperatures and high pressures. It consists of a cantilever beam balance insert for the BYU High Pressure Controlled Profile (HPCP) reactor. The balance unit measures the mass loss of the particles as they oxidize in the HPCP reactor. It includes a force transducer, a ceramic cantilever beam, and a platinum wire-mesh sample basket. The basket is secured to the cantilever beam and extends into the reactor tube through one of the optical access ports of the HPCP reactor. This facility will permit measurements of the oxidation and gasification rates of char particles at pressures up to 17 atm and temperatures up to 1700 K. Bateman et. al., (1994a) successfully demonstrated the use of this experimental setup to study the effect of pressure on oxidation rates of mm-sized char particles. Bateman et. al., (1994b) conducted experiments with Utah bituminous and North Dakota lignite coals at 1, 5, and 7.5 atm total pressure. In these experiments, 5 and 8 mm diameter particles were used. The reactor temperature was varied between 900 and 1200 K and the bulk gas was air. Following the ignition and devolatilization stages, continuous char oxidation rates were measured to burnout. The oxidation rates were significantly increased with increased pressure but found to be independent of temperature. The effects of particle mass and pressure on the oxidation rates of a Utah bituminous char particle are shown in Figures 2 and 3, respectively. The effect of multiple particles on the oxidation rates was also studied. A pair of similarly sized individual particles of coal had similar oxidation times to a single particle having a mass equal to the sum of

the pair. The observed pressure effects could not be explained by a simple global model based on oxygen diffusion to the surface.

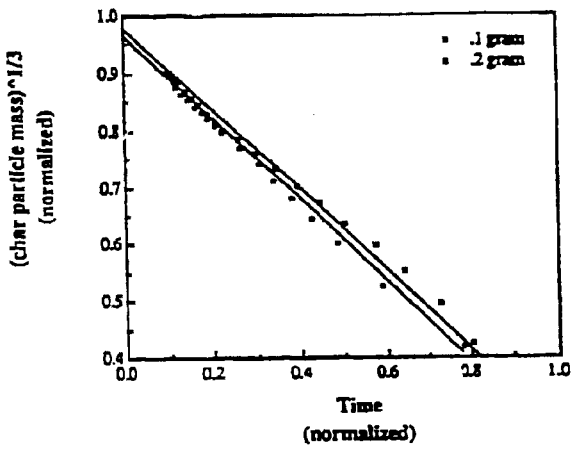


Figure 2. Normalization of the Oxidation Curve for Utah Coal, Sizes 0.1 Gram and 0.2 Gram, in the HPCP Reactor for a Gas Temperature of 900 K and an Air Flow Reynolds Number of 126 at Atmospheric Pressure.

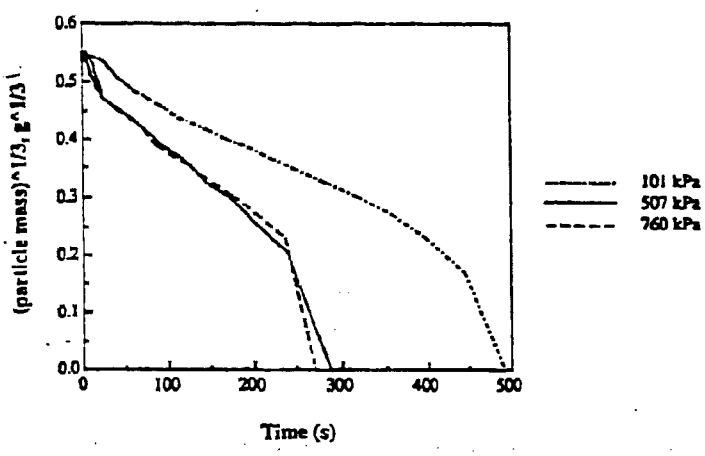


Figure 3. Comparison of Trends of 101 kPa, 507 kPa, and 760 kPa Tests for Utah Coal, Size 0.2 gram, in the HPCP Reactor for a Gas Temperature of 900 K and an Air Flow Re=126.

This study provided the first kinetic rate data for large coal particles at high pressures. The data gave significant new insights into the oxidation process regarding the temperature and pressure dependence and the role of the ash layer. The results also raised important questions: (1) What processes cause the significant effects of both temperature and pressure? (2) Why does the ash layer contract as it does during burning? (3) Why does a group of smaller particles burn like one large particle? (4) Why does the rate of burnout accelerate toward the very end of burnout? (5) Why do some low rank particles ignite before devolatilization? and (6) Why do some bituminous coal particles explode during devolatilization? Additional analysis and experiments will be conducted to answer these questions and develop a new large particle oxidation submodel.

Advanced Fixed-Bed Model Development and Evaluation (BYU) - The objectives of this task are to develop an advanced fixed-bed model incorporating the advanced submodels for large particle devolatilization and large particle oxidation and gasification, and to evaluate the model by comparing its predictions with the experimental data available from the literature. Advanced fixed-bed gasifiers are an integral part of the U.S. Clean Coal Technology (CCT) program and have been considered for integrated gasification combined cycle (IGCC) power generation systems and for mild gasification of lump coal. Recently, novel configurations have been proposed for gasification of caking coals and for mild gasification of lump coal. These configurations include the staged PyGas gasifier of the Gasification Product Improvement Facility (GPIF) project (Brown and Sadowski, 1991), and rotary kiln gasifier of the ENCOAL project (USDOE, 1993). Most of the available fixed-bed models can simulate only traditional fixed beds and there is a need to develop an advanced fixed-bed model capable of simulating both the traditional and emerging, novel gasifiers.

In a previous DOE-METC project, a one-dimensional model for fixed-bed combustion and gasification of coal, FBED-1, was developed (Radulovic et al., 1994). The FBED-1 model has achieved major advances in the treatment of various chemical and physical processes as well as in the solution procedure and has been extensively evaluated. There are still several important issues which must be resolved in the development of an advanced fixed-bed model capable of providing realistic simulations of the emerging configurations. These issues include treatment of large particle devolatilization processes, large particle gasification and oxidation processes, development of robust numerical solution methods, treatment of multiple additions and withdrawals of gases, handling of different flow patterns such as cocurrent, crosscurrent and countercurrent, and evaluation of the advanced model. The FBED-1 model provides a solid foundation for further developments which are continuing in this laboratory.

In order to simulate the combined-bed gasifiers, a preliminary integration of the cocurrent flow option was implemented in the FBED-1 model. The modified code was successfully used to simulate the PyGas gasifier of the GPIF project. The pyrolyzer section was simulated using the zero-dimensional submodel, FBED-0. The freeboard section was simulated by assuming full chemical and thermal equilibrium for the gas phase and no heat and mass transfer between the gas and solid phases. Both the upper and the lower fixed beds were simulated using the modified, fixed-bed model. The detailed profiles for the countercurrent and cocurrent sections of the gasifier are shown in Figure 4. The results predicted about 10% conversion of feed coal in the cocurrent section of the gasifier. Char consumption is due to gasification by CO_2 and H_2O. The gasification rates are higher at the top of the cocurrent section where temperature is relatively high. These rates decay as the solids cool down due to the endothermic gasification reactions. H_2O shows considerable depletion whereas the amounts of CO_2 remain almost constant. Only small amounts of CH_4 are produced in this section. The amounts of CH_4 produced in the countercurrent section are slighily larger. Large amounts of CO and H_2 are produced in the countercurrent section. The peak solid temperature is about 1500 K which is desirable for dry-ash gasifiers such as the PyGas staged gasifier. The product gas, which is the mixed stream from the cocurrent and countercurrent sections, is rich in CO and H_2, and has small amounts of CH_4.

Additional improvements will be made in the FBED-1 model to make it a design and analysis tool of significant industrial utility. These improvements will include incorporation of a large particle devolatilization submodel and a large particle oxidation submodel being developed under this project. A robust solution method suitable for stiff, non-linear problem of fixed-bed systems will be implemented. The model will be evaluated by comparison with the experimental data.

Submodel for Lean Premixed Combustion of Natural Gas in Industrial Gas Turbines (BYU) - A submodel for lean, premixed combustion (LPC) of natural gas in industrial gas turbines is being developed and implemented in a model being developed under independent funding for practical combustors. The submodel must incorporate finite-rate chemistry and predict NO_x and CO emissions at conditions approaching blowout (0.5 to 0.6 stoichiometric ratio in the primary zone). Proper accounting of the effects of chemistry/turbulence interactions is important. The Monte Carlo pdf method (Pope, 1985) is being considered as a basis for the submodel. A partially-stirred reactor (PaSR) model is being used to investigate the Monte Carlo pdf method. The PaSR (Chen, 1993) is similar to the perfectly stirred reactor (PSR) model, with no spatial variation of mean fluid properties. The difference

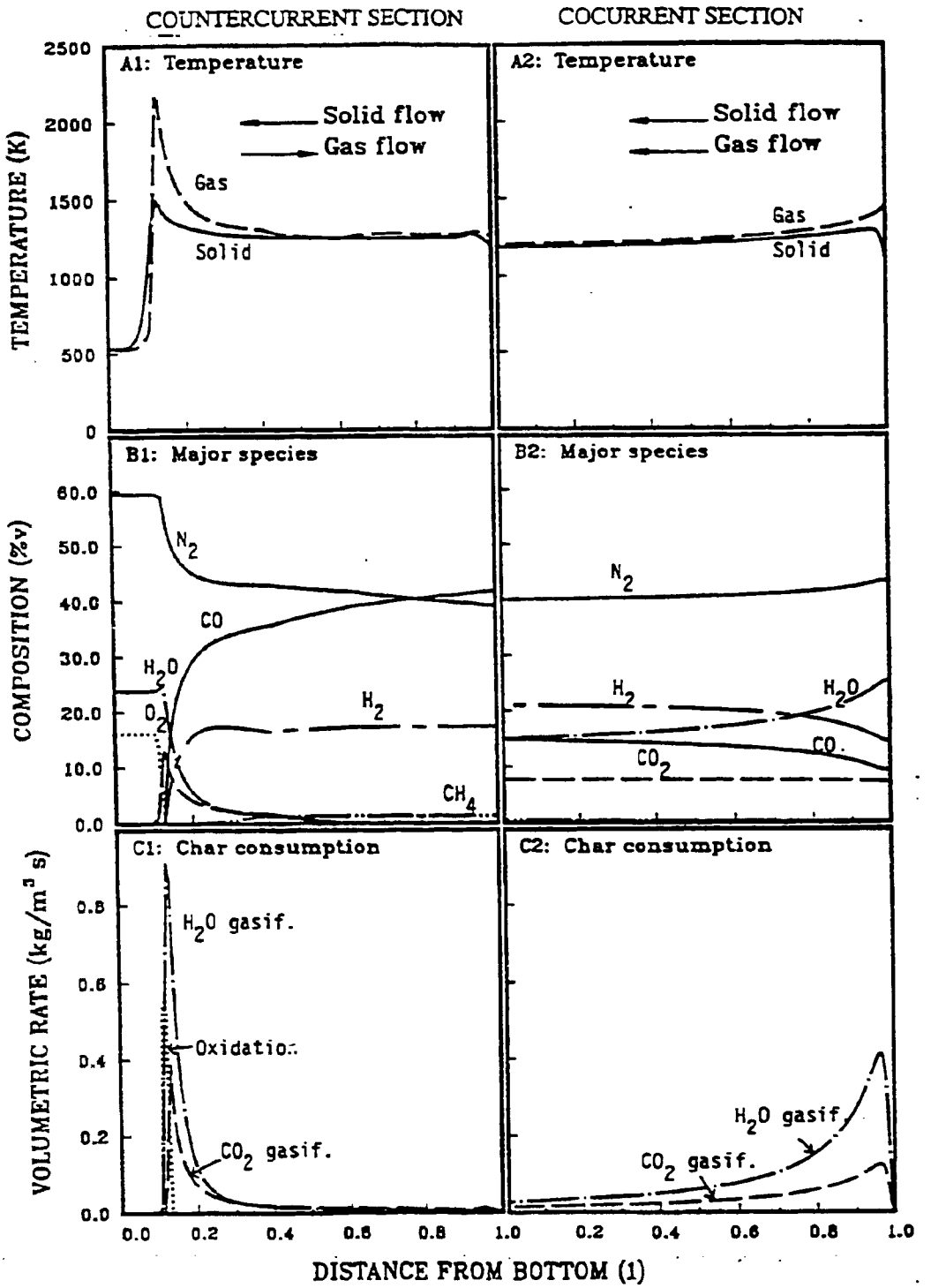


Figure 4. FBED-1 Predictions for the Countercurrent and Cocurrent Sections of the Staged PyGas Gasifier

is that molecular mixing is incomplete, so that the reactive fluids are not completely diffused into one another. Hence, there is a finite rate, given by a mixing frequency, at which the reactive fluids are mixed on a molecular level, as well as a finite rate at which they react after being mixed. Both the mixing and reaction time scales are important in the PaSR.

Calculations are being performed at conditions typical of LPC in stationary gas turbine combustors (premixed methane in air, 700 K inlet, 0.5~0.6 equivalence ratio, 15 atm, 10-20 ms residence time, and 100-5000 s^{-1} mixing frequency). The PaSR is being studied for several reasons. First, it provides an opportunity to understand the Monte Carlo pdf method in a simple model framework without the added complexities of fluid dynamics calculations. Second, many of the subroutines in the PaSR code may be applicable to a pdf submodel in PCGC-3 with little or no modification. Third, the PaSR model is a viable engineering model in its own right, with capability for including full chemistry (critical for predicting trace species). Fourth, the lessons learned from the PaSR are directly relevant to pdf methods for multi-dimensional flow, since the pdf equation for the PaSR is a degenerate case (integrated over reactor volume) of the pdf for the multi-dimensional case. Fifth, the PaSR provides an alternate test bed (to the more commonly used stirred-tank and plug-flow models) for evaluating reduced chemical schemes. And sixth, the PaSR provides a test bed for mixing models, a critical element of pdf models, in the context of full kinetics.

Results have been obtained using a reduced Miller-Bowman scheme (without C_2 chemistry and prompt NO) (Correa and Braaten, 1993). The effects of mixing rate, pressure, and mixing model were investigated. The effects of mixing rate at 15 atm and using the modified Curl's mixing model (Chen, 1993) are shown in Fig. 5. The importance of in-combustor mixing, even for premixed reactants, is clearly shown. Mixing of hot, partially-reacted particles in the combustor with the cold, incoming particles, is necessary to sustain combustion. Stable combustion was not achievable with a mixing frequency of 300 s^{-1} and lower. As shown in the figure, the PaSR converged to the non-reacting state at a mixing frequency of 300 s^{-1}. The higher the mixing frequency, the higher the temperature and the more complete the combustion, as the cold particles are quickly heated and brought to reaction temperature. The stochastic steady state is reached more rapidly with more rapid mixing. CO and the heat release chemistry (i.e. temperature) converge to the stochastic steady state at about the same rate, while NO converges considerably more slowly. This observation is in agreement with that of Correa and Braaten (1993). Scatter plots and pdf plots (not shown here) showed that the particles became more uniform in their properties as mixing frequency increased.

The above results were obtained with the modified Curl mixing model (Chen, 1993). The effect of the mixing model was investigated by repeating the calculations for a mixing frequency of 1000 s^{-1} with the Interaction-by-Exchange-with-the-Mean (IEM) model, but the solution converged to the non-reacting case. It was therefore decided to reproduce the results of Correa (1993) for CO/H_2/air combustion with a goal of determining whether the IEM model is implemented correctly in the PASR code. These calculations are in progress. If successful, calculations will then be performed with a full kinetic scheme for natural gas combustion, with a view toward obtaining a suitable reduced mechanism for use in a submodel to be implemented in a 3-D gaseous combustion code for application to lean, pre-mixed gas turbines.

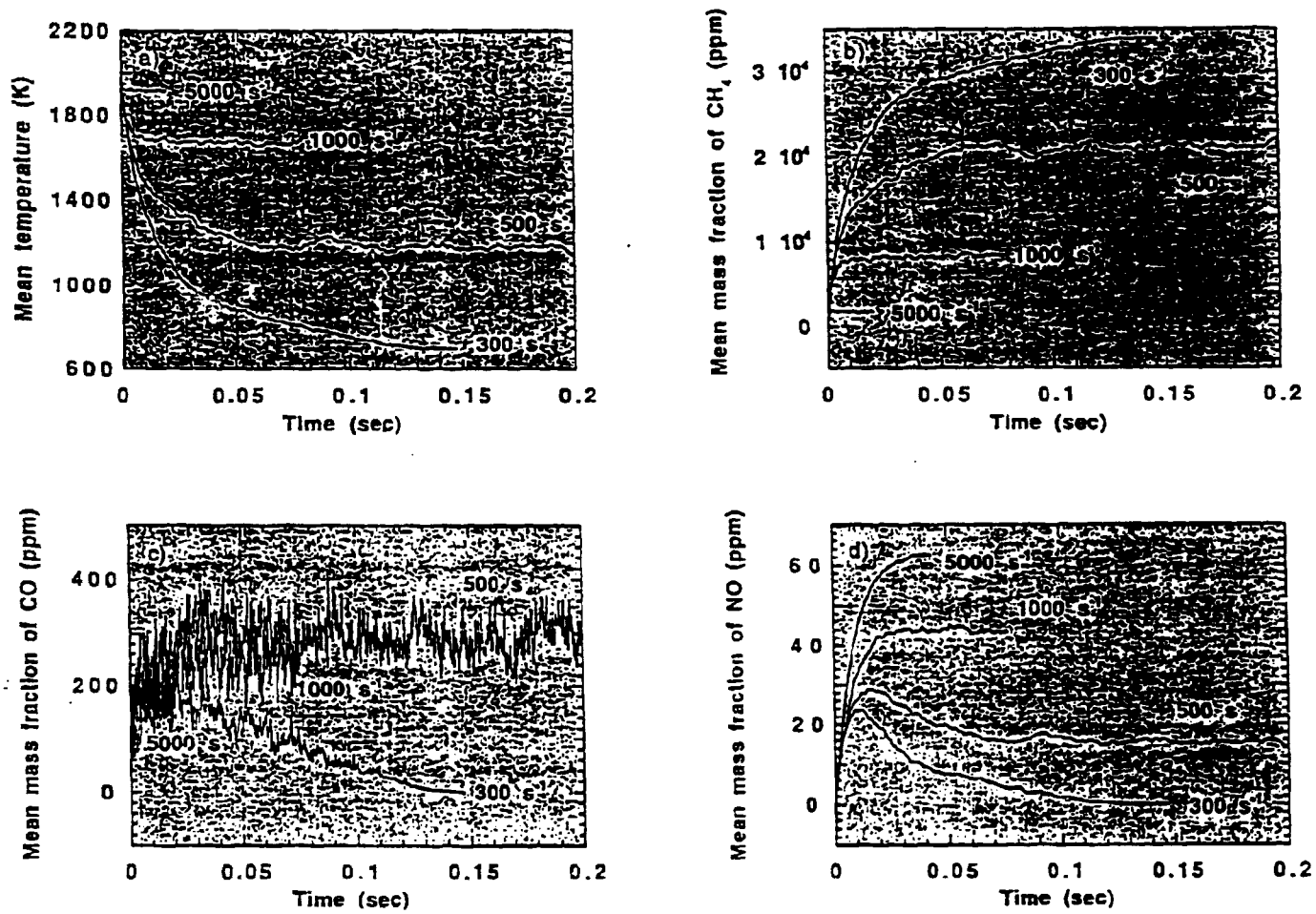


Figure 5. PaSR Predictions for Fuel-Lean, Premixed Methane Combustion in Air (0.6 Equivalence Ratio, 15 atm, 700 K Inlet, 10 ms Residence Time

FUTURE WORK

Work during the next year will include: 1) completion of simplified FG-DVC models for fixed-beds and fluidized-bed reactors; 2) integration of a particle heat transfer model with a simplified FG-DVC model; 3) complete stochastic particle dispersion model; 4) complete formulation of transport reactor code; 5) complete integration of simplified FG-DVC model into fluidized-bed code; 6) complete formulation of a large particle oxidation model; 7) complete integration of a large particle devolatilization submodel (based on FG-DVC) into FBED-1; 8) implement improved numerical solution method for FBED-1; 9) extend the PaSR pdf model to the case of natural gas combustion and use the model to develop/evaluate a suitably reduced kinetic mechanism for application in multi-dimensional flame codes.

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