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ADVANCED PULVERIZED-COAL COMBUSTOR FOR
CONTROL OF NO_x EMISSIONS

Second Quarterly Report
For the period
25 December 1980 through 3 April 1981

R. L. Pam, S. T. Suttman, J. T. Kelly, E. K. Chu

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Prepared For

U.S. Department of Energy
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ABSTRACT

Second quarter (25 December 1980 - 3 April 1981) results under the Advanced Pulverized Coal Combustor for Control of NO_x Emissions Program (DOE Contract DE-AC22-80PC30296) are reported. The program uses an idealized one-dimensional combustor, incorporating fuel staging. A comprehensive computer model is being developed under the program to allow NO mechanisms and rates to be extracted from the experimental data.

During the quarter, modification of the existing PROF code to include particle effects was begun. Various aspects of the solution procedure are described in this report. Refinements in the kinetic thermochemical data are completed, providing satisfactory agreement between calculated NO predictions and selected experimental data.

Combustor mechanical design was described last quarter. The heat transfer analysis used in the design of the combustor is reported herein. Initial combustion tests will be run next quarter.

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1. CONTRACT OBJECTIVES

The objective of the proposed program is to quantify and define, in terms of the kinetic mechanisms and rates, the pulverized coal NO_x emission control benefits of fuel staging. A well defined experimental combustor, incorporating fuel staging, will be built and a comprehensive computer model constructed to allow mechanisms and rates to be extracted from the experimental data. Experiments are proposed in which fuel and air injection parameters will be varied to determine the effect of first and second stage fuel loading ratio on NO_x emissions and combustion.

The computer analysis of the resulting data will yield quasi-global kinetic rates and mechanisms for the process. These rates can ultimately be used in a realistic engineering model to help guide industrial combustor designers.

The characterization of the fuel staging NO_x reduction process in pulverized coal combustion will allow more flexibility in designing low NO_x combustors. The increased flexibility will help overcome industrial combustor configurational, volumetric, and material constraints limiting present NO_x control techniques.

2. CONTRACT TASKS

During the first quarter of the contract, the combustor model governing equations and gas phase NO_x kinetics were formulated and combustor design was completed. In the second quarter, incorporations of governing equations into the PROF code was begun, as was combustor fabrication and installation. Progress is described in the following paragraphs.

2.1 ANALYTICAL MODEL DEVELOPMENT (TASK 1.1.2)

The objective of this task is to create a comprehensive data analysis tool to develop pulverized coal combustion and NO_x formation kinetic mechanisms and rates from the idealized combustor data.

During the first two quarters of this program, the computer model used as the base for the comprehensive data analysis model was defined and the modifications needed to upgrade the base model were outlined. The Acurex development PROF (Pre-Mixed One Dimensional Flame) computer code was selected as the base model, since this code satisfies the detailed treatment of chemical and physical events and includes upstream heat and mass transfer required of the comprehensive model (reference 1). Progress made in this quarter toward incorporating particle effects into the PROF code is described in the following section.

2.1.1 INCORPORATING PARTICLE EFFECTS INTO THE PROF CODE

The governing equations for a particle-gas system were developed in this quarter of the program (reference 2). They are repeated here:

- Gas phase enthalpy balance,

$$\begin{aligned} \dot{m} \frac{dh}{dx} = & A\dot{Q} - \frac{d}{dx} \left(\sum_i A J_i h + A k \frac{dT}{dx} - C_w \dot{Q}_w \right) \\ & - A \sum_j Q_{c_j} - \sum_j \frac{d\dot{m}_{p_j}}{dx} (h_{p_j} - h) \end{aligned} \quad (1)$$

- Gas phase species balance for each specie i ,

$$\begin{aligned} \dot{m} \frac{dY_i}{dx} = & A W_i - \frac{d(A J_i)}{dx} - C_w J_{W_i} \\ & - \sum_j \left[\frac{d\dot{m}_{p_j}}{dx} (Y_{p_{ij}} - Y_i) - A J_{p_{ij}} \right] \end{aligned} \quad (2)$$

- Particle enthalpy balance for each particle group j,

$$\dot{m}_{p_j} \frac{dh_{p_j}^*}{dx} = \dot{Q}_{c_j} A + \frac{d\dot{m}_{p_j}}{dx} (h_{p_j} - h_{p_j}^*) + \dot{Q}_{r_j} A \quad (3)$$

- Particle mass balance for each particle group j,

$$\frac{d\dot{m}_{p_j}}{dx} = \dot{r}_j''' \quad (4)$$

(See table 1 for a list of symbols.)

These equations are rewritten in linear finite differenced form;

$$\left[(\text{at grid step } n) \, ds = \left[\frac{(S_{n+1} - S_n) + (S_n - S_{n-1}))}{2} \right] \right] \quad (5)$$

S_n represents an axial position in the combustor and S_{n+1} , a position upstream and S_{n-1} , a position downstream. Using the modified distance coordinate $\bar{s}$, $d\bar{s} = \left(\frac{A_0 ds}{A_p T^{1.659}} \right)$, the equations have the correct form for input into the PROF code (reference 1).

In the original PROF code, a conservation equation error vector, RHS, is generated at each grid step at each iteration. This matrix is extended and modified to include the effects of different particle groups as follows:

$$\begin{aligned} \text{RHS (1)} &= H_{\text{gas in}} - H_{\text{gas out}} \\ &- (\text{heat loss from gas to particles}) \\ &- (\text{heat loss to wall}) \\ &- (\text{volumetric heat loss}) \\ &+ (\text{heat gain from particles to gas}) \end{aligned} \quad (6)$$

$$\text{RHS (2)} = P_{\text{in}} - \sum_i P_i \quad (7)$$

$$\begin{aligned}
\text{RHS (I+2)} &= P_i \text{ in} - P_i \text{ out} \\
&+ (i \text{ formed from gas reactions}) \\
&+ (i \text{ lost to walls}) \\
&+ (i \text{ formed from particle reactions})
\end{aligned} \tag{8}$$

where I = 1, IS (total number of species)

$$\text{RHS (IS + 2 + J)} = \dot{m}_{p_{j \text{ in}}} - \dot{m}_{p_{j \text{ out}}} - \dot{r}_j' \tag{9}$$

where J = 1, IP (total number particle of groups)

$$\begin{aligned}
\text{RHS (IS + 2 + IP + J)} &= H_{p_{j \text{ in}}} - H_{p_{j \text{ out}}} \\
&- (\text{heat loss from particle group } j \text{ to gas}) \\
&+ (\text{heat gain from gas to particle group } j)
\end{aligned} \tag{10}$$

where J = 1, IP (total number particle of groups)

With the combustor inlet conditions specified and first guesses for the iteration variables chosen, the RHS matrix terms are calculated. The iteration variables are gas temperature, T, PM (pressure x gas molecular weight), and the partial pressure of each gas specie, P_i as in the original PROF code, with the addition to the iteration variables of the mass flow rate of each particle group, $\dot{m}_{p_j}$, and the temperature of each particle group, T_{p_i} . The RHS terms are not equal to zero with most first guesses; instead, there is some error associated with each RHS expression, i.e., $\text{RHS (1)} = \Delta E (1)$. When all ΔE 's are equal to zero, the corresponding values of the iteration variables represent the solution to the conservation equations.⁽¹⁻⁴⁾ To find this solution, the A matrix is formed, which represents the derivatives of the conservation equations with respect to the natural log of the iteration variables as shown in

figure 1. When the A matrix is inverted and multiplied by ΔE ($\Delta E = \text{RHS}$), ΔV is obtained:

$$\text{RHS} \times \left[\underline{A} \right]^{-1} = \Delta V \quad (11)$$

The new guess for the iteration variables is, therefore, given by:

$$V_{\text{new}} = V_{\text{old}} + \Delta V. \quad (12)$$

Here, ΔV is a vector which represents the change in the iteration variables required to make the RHS vector equal to zero. V_{new} , then is a vector of the new iteration variable values to be used as first guesses. Successive iterations, if properly constrained, will drive ΔE to zero.

The preceding RHS equations and the derivative terms in the A matrix include a term representing the mass fraction of specie i at the particle surface of particle group j , $Y_{p_{ij}}$. This quantity is determined from a specie balance at the particle surface:

$$\rho_e U_e C_m \left(Y_{p_{ij}} - Y_i \right) - \dot{r}_j^{'''} Y_{p_{ij}} + \sum_m \left(\mu_{m_i}^P - \mu_{m_i}^R \right) W_{p_{mj}} = 0 \quad (13)$$

This equation represents the sum of the rate of reaction of the specie on the particle surface, the rate of diffusion of the specie to the surface and the transport of the specie away from the surface with the mass loss rate from the coal particle.

The total mass loss rate from the particle group j , $\dot{r}_j^{'''}$, in equations 9 and 13 is equal to the mass loss due to devolatilization plus the mass loss due to surface oxidation of the coal;

$$\dot{r}_j^{'''} = R_{V_j} + R_{C_j}. \quad (14)$$

Conservation Equations	$\partial \ln P_i$			$\partial \ln \dot{m}_{p_j}$		$\partial \ln T_{p_j}$	
	$\partial \ln T$	$\partial \ln (PM)$	$i = 1, IS$	$j = 1, IP$		$j = 1, IP$	
$(PM) h_{gas}$ P P_i $(i = 1, IS)$	Original PROF A Matrix						
$\dot{m}_{p_j} (j = 1, IP)$ $h_{p_j} (j = 1, IP)$							
				Modified Prof A Matrix -- Particle Effects Added			

Figure 1. Form of A Matrix

The devolatilization reaction rates are of the form

$$R_{V_j} = k_{V_j} (V_{\infty} - V) . \quad (15)$$

$$\text{where } k_{V_j} = A_v \exp \left[- \frac{E_v}{RT_{p_j}} \right]$$

V_{∞} = amount volatilized at $t = \infty$

V = amount volatilized at t

This form associates different rate constants with the devolatilization of each volatile specie.

The surface reaction rates are of the form,

$$R_{C_j} = k_{C_j} P_{p_{i_j}} A_{p_j}$$

$$\text{where } k_{C_j} = A_c \exp \left[- \frac{E_c}{RT_{p_j}} \right]$$

$P_{p_{i_j}}$ = partial pressure of reactant specie i at particle group j surface

A_{p_j} = surface area particle group j

This form also associates a different temperature-dependent rate constant with each surface reaction. With experimentally determined reaction rate data, the rates of devolatilization and surface reaction will be accurately modeled in the modified PROF code.

The complete form of the RHS vector and A matrix incorporating the particle effects mentioned above has been completed, written in Fortran, and organized into separate subroutines to be added to the PROF code.

The particle subroutines are named PARINP, PARKIN, and PPROP. Subroutine PARINP is added to the main program ACEF (see reference 1) after subroutine READIN. READIN reads in gas species names, initial

relative moles, and guessed final relative moles. Subroutine PARINP reads in inlet particle properties including the number of particle groups, IP; the diameter, DJ; the number density, NJ; the mass flow rate, MPIN, of each particle group; the number of volatile reactions, IVR; the total number of particle reactions, IR; the heat of formation, HFOP; and heat capacity, CPC, of the coal particles. This subroutine also reads in the first guesses of properties at a given grid step, including the outlet temperature, T_p , and flow rate, MPOT, for each particle group. PARINP calculates the input and first guess outlet enthalpy of the coal based on the temperatures read in.

Subroutine PARKIN is called after subroutine PARINP in ACEF to read in the particle kinetic reaction data for the volatile and surface oxidation reactions. This data includes the energy of activation and the pre-exponential factor for each reaction. PARKIN also records the stoichiometric coefficients for each specie I in each particle reaction J, in matrix STO (J,I).

The main program, ACEF, then calls subroutine OBTAIN which obtains the molecular weight and thermochemical properties for each gas specie. Then, subroutine EPROP is called at the first grid step, which provides the inlet conditions. Within EPROP, subroutine FLAME is called to calculate the input gas specie thermodynamic properties. These properties are returned to ACEF. ACEF then calls EPROP again to calculate the properties at each grid step based on the output first guesses. Subroutine FLAME is called to calculate the thermodynamic properties and the A and RHS matrices for gaseous combustion at each grid step. Subroutine PPROP is called within FLAME to calculate all the particle properties and derivatives needed to add to A and RHS. PPROP returns to

FLAME just prior to the inclusion of the gas phase chemical-kinetic rate terms. When FLAME converges to the solution at a grid step, the correct variable values are returned to ACEF and the procedure repeated for the next step.

The new particle subroutines, PARINP, PARKIN, and PPROP are written generally to include any number of volatile reactions, any number of surface oxidation reactions, any number of surface oxidation reactions and any number of particle groups.

In the next quarter, the particle radiation model will be added to the particle subroutines. The new subroutines will be debugged and verified with limit data. Specific volatile and surface reactions and the corresponding kinetic data will also be added. New gas species formed from particle devolatilization will be added along with kinetic and thermodynamic data for these species.

The overall operation of the code will be tested by running different modeling options (well stirred, plug flow, flame, etc.) under various conditions of temperature of stoichiometry to check solution validity and convergence.

2.1.2 KINETIC MODELING (TASK 1.1.1)

As reported in the monthly reports, the elementary reaction set (adapted from that of Energy and Environmental Research Corporation) for hydrocarbon and nitrogen species has been refined during the program second quarter. Initial computations of nitrogen species production showed some discrepancies in comparison to selected experimental stirred reactor data. Differences in the EER and Acurex thermochemical data (taken from JANAF curve fits) were noted for the heat of formation at 298K

Table 1. List of Symbols

A_o	Reference combustor cross sectional area	cm^2
A	combustor cross sectional area	cm^2
A_p	particle surface area	cm^2
C_p	specific heat	cal/gm
d_j	particle diameter	cm
h	enthalpy of bulk gas	cal/gm
h_i	enthalpy of bulk species i	cal/gm
h_{p_j}	gas enthalpy at particle j surface	cal/gm
$h_{p_j}^*$	condensed phase enthalpy of particle j	cal/gm
J_i	axial species flus	gm/sec cm^3
$j_{p_{ij}}$	species flux to particles	gm/sec cm^3
J_w	wall species flux	gm/sec cm^2
k	thermal conductivity of bulk gas	cal/sec cm K
$\dot{m}$	mass rate of gas	gm/sec
$\dot{m}_{p_j}$	mass rate of particles j	gm/sec
n_j	number of particles per volume	cm^{-3}
P_i	partial pressure of specie i in bulk gas	atm
$P_{p_{ij}}$	partial pressure of specie i at partice j surface	atm

Table 1. (Continued)

PM	pressure x molecular weight	atm-gm/mole
$\rho_e U_e c_m$	mass transfer coefficient	gm/sec
ρ	gas density	gm/cm ³
$\dot{Q}$	gas volumetric heat loss	cal/cm ³ sec
$\dot{Q}_{c_j}$	particle/gas heat transfer	cal/cm ³ sec
$\dot{Q}_{r_j}$	particle to particle radiative transfer	cal/cm ³ sec
$\dot{Q}_w$	combustor wall heat transfer	cal/cm ² sec
$\dot{r}_j'''$	mass rate of gas from particles	gm/sec
$\dot{R}_c''$	heterogeneous oxidation rate	g/cm ² -sec
$\dot{R}_v$	devolatilization rate	g/sec
$\dot{R}_w$	moisture vaporization rate	g/cm ² -sec
s	axial distance	cm
$\bar{s}$	modified axial distance	cm ⁴ /gm-(°K) ^{1.659}
T	temperature of gas	K
T_{p_j}	temperature of particle	K
W_i	gas phase reaction contribution to species i	gm/cm ³ sec

Table 1. (Concluded)

$w_{p_m j}$	particle reaction rate of mth reaction on particle j surface	gm/cm ³ sec
x	axial distance	cm
y_i	mass fraction of species i	
$\nu_{m_i}^p$	stoichiometric coefficients for product specie i in particle reaction m	
$\nu_{m_i}^R$	stoichiometric coefficients for reactant specie i in particle reaction m	
$y_{p_i j}$	mass fraction of species i at surface of particle j	

and specific heat values for a few species. All the species compared are listed in table 2. The percentage differences relative to the data used by EER are tabulated for the temperature range of interest ($\approx 1000\text{K}$ to 2500K) for the species whose discrepancies are greater than 1 percent.

The sensitivity of the computational scheme to the curvefit values has been evaluated by eliminating the discrepancies of table 2 and rerunning of sample data comparisons. Two of the comparisons are shown in figures 2 and 3. Although the discrepancies were found to be small (less than 10 percent), the effects on NO predictions are significant — especially on the fuel-lean side. With the updated thermochemical set, higher NO conversion was predicted than that of the old thermochemical set under fuel-lean conditions, resulting in overpredicting the experimental data. Under fuel-rich conditions, the same trend was observed except that satisfactory agreement with the experimental data was retained. The overprediction of NO was found to be particularly sensitive to the heat of formation of CHO. Since varying the heat of formation of any species can result in variations in the equilibrium constant and, therefore, the reverse reaction rate, adjustments to the rate constants of all reactions containing CHO may be required to further improve the comparisons. This level of refinement is not warranted at this time. Although discrepancies remain, the new set of thermochemical data gives closer comparisons of NO under fuel-rich conditions for the retention of the added NO case (figure 3).

2.2 COMBUSTOR DESIGN AND FABRICATION (TASK 1.2)

This quarter, fabrication of combustor parts was completed and installation was begun. Combustion tests will be initiated next quarter.

TABLE 2. Comparison of EER and JANAF Thermochemical Data

Species	Heat of formation at 298K			Specific heat (percent)
	EER (Kcal/mole)	JANAF curve fit (Kcal/mole)	Percent	
CO	-26.4165	-26.417		5.0
CO ₂	-94.054	-94.054		
CH ₄	-17.895	-17.895		
H ₂ O	-57.7979	-57.798		
O ₂	0	0		
N ₂	0	0.79×10^{-3}	X	
H ₂	0	0.2×10^{-3}	X	
H	52.10	52.1		
O	59.559	59.559		
N	113.0	112.96		
NO	21.58	21.58		2.0
HO	9.492	9.432	1.0	
HO ₂	5.0	5.0		
CH ₃	34.82	34.82		
CH ₂ O	-25.95	-27.70	7.0	
CHO	9.0	-2.9	132.0	
NO ₂	7.91	7.91		
N ₂ O	19.61	19.61		
CH ₂	92.35	92.07		
CH	142.0	142.0		
HNO	24.5	23.8	3.0	7.0
H ₂ N	40.07	40.07		
H ₃ N	-10.97	-10.97		
HN	90.0	81.0	10.0	
CN	104.0	104.0		
CHN	32.3	32.3

Figure 2. Conversion of HN_3 to NO_x — 1300 ppm Addition NH_3

Figure 3. Retention of Added NO — NO Addition

In the last Technical Progress Report (reference 2), the combustor components and subsystem mechanical design was described. The heat transfer analysis used in the design of the combustor is described below.

Thermal Design

The furnace design had three objectives:

1. Achieve necessary residence times for staging
2. Maintain exterior shell temperatures below $\sim 250^{\circ}\text{F}$
3. Achieve a reasonable time/temperature profile, simulating normal combustor profiles.

Typical time/temperature profiles are shown in figure 4. By matching volumetric firing rates (Btu/hr-ft^3) and heat loss between full- and sub-scale furnances, the desired time/temperature profile was translated to a desired temperature profile along the subscale combustor length. Various combinations of combustor length with 8- and 6-inch diameters were tried, with the 8-inch diameter being selected to provide lower flow velocities and shorter combustor lengths. Having chosen these major parameters, a combustor heat analysis was done to define insulation and backfiring requirements to achieve the proper heat loss.

Similar furnaces used by other researchers (references 3 and 4) were known to have excess heat loss, resulting in temperature profiles below the desired levels. Therefore, a backfiring system was devised to provide control of the losses. The result is a main combustor surrounded by an annular combustor for backfiring (figure 5).

Heat transfer

The heat transfer situation is a complex one due to the interaction of the two firing systems and the variations of gas and wall temperatures in the flow direction. Pershing had developed a computer model

Figure 4. Relation of HVT Furnace Gas Temperature to Height Above Top of Burners⁵

Figure 5. Combustor Cross Section

(reference 3) that handled a simple combustor, i.e. the main firing system only. This model was revised to include the annular backfiring, assuming that heat transfers outward only.

Calculation procedure:

1. Assume T_b and $T_{b,0}$ (code calculates h) calculate change in T_b and $T_{b,0}$ along the length using finite element analysis. Size outer refractory to keep $T_s < 250^\circ\text{F}$.
2. Set calculated $T_{b,0}$ distribution as the boundary condition for the inner refractory lining. Assume no heat transfer across backfiring region.
3. Set T_{af} and T_i and do finite element analysis to achieve inner time/temperature history.
4. Check heat flow assumption across the backfire zone. If far from adiabatic, estimate effects on temperature profile.
5. Iterate, varying geometries, gas temperatures, and wall temperatures (most appropriate values were selected based on data of similar furnaces).

Results

Figure 6 shows the backfire gas and coal products temperature profiles for several inlet temperatures. The effects of co-flow and counterflow backfiring are shown. The conditions giving the lowest profile (compared to desired profile) are backfire gas temperature = 2600°F , average coal gas temperature = 2700°F . Flexibility in operation is built in by a backfire burner that can operate from 5 to 80,000 Btu/hr (30,000 through 50,000 is anticipated) and 0 to ~70 percent excess air. This firing rate should compensate for the fact that the final design of the backfiring system is not truly annular.

Figure 6. Temperature Versus Height Profiles - Subscale Combustor

3. WORK PLANNED FOR NEXT REPORTING PERIOD

In the next quarter, initial runs of the updated PROF code will be made, incorporating particle effects, updated thermochemical data and a radiation model. Fine-tuning to match published experimental data in limiting cases will begin.

The combustor will be completed, and testing initiated. A shakedown period on gas and then coal will verify mass balances, and then advanced air staging tests will be initiated.

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