DOE/PC/94254--T3

**Computational Modeling and Experimental Studies on NO_x Reduction
Under Pulverized Coal Combustion Conditions**

Technical Progress Report
Third Quarter

July 1, 1995 - September 30, 1995

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Work Performed Under the Contract DE-FG22-95PC94254

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INTRODUCTION

An experimental plan outlining the first year's activity was sent to Dr. Lori Gould, Project Officer/ Contracting Officer's Technical Representative on April 24, 1995. An approval was received with some questions on June 15, 1995. However, with some foresight of the director of the in-house combustion group of the PETC, Dr. Ekmann, a tentative hold-off on the purchase of the equipment was requested by the project officer on June 29, 1995. Enclosed with that request were some of Dr. Ekmann's concerns. The research team spent the month of July in study of pertinent literature as well as in the preparation of the responses to Dr. Gould's comments and Dr. Ekmann's concerns. These responses included the choice of the reactor, reactor design, rate of gas heating, detailed test matrix and answers to host of other comments. Upon review of the above information submitted on July 24, 1995 by the Rust research team, the project officer called for a conference call on September 6, 1995 which involved the PI (Dr. Kumpaty), the research consultant (Mr. Subramanian), Dr. Gould and Dr. Ekmann. Except for the furnace and the reactor, other pieces of equipment were approved for purchase. An important issue was to make sure the Lyndberg could accommodate the measurement of wall temperature of the reactor as well as at least the exit temperature of the gas mixture inside the reactor. Dr. Ekmann insisted that further calculations be made on the rate of gas heating without taking radiation into account. Accordingly, calculations pertaining to the rate of gas heating based on convection were performed and submitted to Dr. Ekmann on September 13, 1995. Towards the end of the

quarter, some items have been ordered such as the Nox analyzer from California Analytical Instruments and some supplies/accessories from McMaster-Carr. Some preliminary computational effort was made on the simulation of the burnout zone during the month of August; however, due to the urgency of the hour, focus was turned completely toward the experimental. This report contains the information emerged through the dialogue between the Rust College research team and the PETC represented by Dr. Gould and Dr. Ekmann during this quarter.

CHOICE OF REACTOR

The reactor chosen is a plug flow tubular reactor, 1.885 cm ID, 65 cm length, assuming straight line velocity and temperature profiles.

What other reactors are considered ? Why rejected?

Many experimental techniques including shock tubes, stirred reactors, turbulent reactors have been considered. Because of physical equipment and processes involved, each of the above had its own merits and de-merits. Shock tubes are accurate with temperature and reaction time determinations but the actual reaction time achievable in a shock tube is very short (1 to 2.5 ms) compared to industrial furnaces. Also, in order to have consistent concentration, the analysis techniques require certain gas volume which may be a constraint with a shock tube. The stirred reactors have the merit of reaction getting to completion but strong back mixing will not be representative of the species found in an actual reburning situation. The actual reburning conditions can be met in large scale turbulent reactors since the dimensional

and kinetic similarity can be maintained. However, there are lot of uncertainty factors such as (laminar or turbulent) nature of the flows, species concentration coupled with mixing influences and profiles of temperature which can not be properly defined and hence meaningful analysis/conclusions can not be arrived at.

Why the experimental setup was chosen

We have chosen to conduct experiments with the laminar flow reactor for the following reasons:

- a) This reactor represents the actual reburning environments.
- b) The experimental conditions are similar to actual industrial furnaces.
- c) The low Reynolds number ($20.6 < 2100$; as shown in the reactor design section) ensures laminar flow, so turbulent effects can be neglected.
- d) The furnace can maintain the reaction temperature for 1100 C and it can reproduce the same temperature for subsequent experiments for comparison.
- e) Other researchers have used some of the design practices proposed and have successfully carried out their experiments. For example,
 - i) J.M. Levy et al (1981) used Helium as diluent gas; used temperatures of 1250-1750 K; used laminar flow tubular reactor; used similar coal (feeding) entrainment media and the same point of injection as ours.
 - ii) S.L. Chen et al (1989) employed $SR1=1.1$; used residence time of 0.4 s; used gas washing method and specific ion electrodes to measure HCN and NH_3 species.

What we hope to simulate with the reactor

We would meter the gases from cylinders through rotameters and mix it in the buffer chamber

so that it represents the flue gas from the primary combustion zone. Then it passes through the reactor where reburning fuel is injected. The NO from combustion zone will be reduced to N₂ and CO₂ through a series of chemical reactions (250 odd reactions). Using this experimental setup we would like to develop data as described under the experiment 1a (See the test matrix).

To prove the device yields information of desired quality

We have proposed to test if CH₄ reburning results in similar NO reduction, as reported by previous researchers.

REACTOR DESIGN

Plug flow tubular ceramic reactor (1.885 cm I.D and 65 cm length).

Central portion of the reactor is enclosed in a 30 cm long, electrically heated laboratory tube furnace (Lyndberg Minimate Model 55035A).

SR2=0.9:

Gas	Volume Flow, cc/min	Density, g/cc	Weight, g/min
CO ₂	321	0.0019768	6.35E-01
O ₂	36.8	0.0014289	5.26E-02
N ₂ /He	1549	1.769E-04	2.74E-01
NO	1.91	0.00134	2.56E-03
CH ₄	41.7	7.167E-04	2.99E-02

Weight of the gas mixture, w_{mix} = 0.9936 g/min.

Average molecular weight of the mixture = $\sum (\% \text{vol (g-mole)} * \text{M.W.}) = 11.392 \text{ g/g-mole}$

Mixture density (273 K, 1 atm) = Av. M.W./Mol. Vol. = $11.392/22.4 \times 10^3 = 5.086 \times 10^{-4}$ g/cc

Volumetric rate of the gas mixture at 273 K = $w_{\text{mix}}/\rho_{\text{mix}} = 1954$ cc/min.

Volumetric flow rate of the gas at 1100 C = $1954 \times 1373/273 = 9826$ cc/min.

A residence time of 0.2 seconds is typical for reburning zone. Therefore, the volume flow in 0.2 seconds is $9826 \times 0.2/60 = 32.75$ cc. For a tubular flow reactor, $(\pi/4) d^2 l = 32.75$ cc.

Assuming 1" O.D. and using Sch. 80 the inside diameter is 0.742", that is 1.885 cm.

Therefore, the length of the reactor where $T=1100$ C is $32.75 \times 4/(\pi \times 1.885^2) = 11.74$ cm.

Lyndberg assures that it can offer its model (55035 A) which has a reactor O.D. of 1" and it promises 1100 C for 12 cm length of reactor. So, we will use the above reactor with 55035A

Lyndberg Minimate furnace.

Central portion of the reactor surrounded by the furnace:

Lyndberg Minimate furnace keeps the 11.75 cm of heated length at 1100 C.

Residence time calculation in the heating zone of the reactor

From the reactor design calculations, we find the volume rate of the gas mixture as 9826 cc/min at 1100 C. For a given reactor cross-sectional area, this would mean $9826/(\pi/4)1.885^2 = 3521$ cm/min of linear velocity. To cover 9.125 cm of reactor length, it would take $9.125 \times 60/3521 = 0.1555$ s.

The heating time of 0.0477 s (See rate of gas heating calculations) is < 0.1555 s. So the gas can easily get heated to 1373 K.

Reynolds Number

Volumetric flow of gas at 1100 C (as in the previous section), $V = 9826 \text{ cc/min}$

Cross-sectional Area of the reactor tube, $A = \pi/4 (1.885\text{E-}02)^2 \text{ m}^2$

Velocity, $v = V/A = 35.21 \text{ m/min} = 58.68 \text{ cm/s}$

Density of the gas mixture at 1100 C, $\rho_{\text{mix}} = w_{\text{mix}} / V_{\text{mix}} = 0.9936/9826 = 1.011\text{E-}04 \text{ g/cc}$

At $SR_2=0.9$

Gas	Percent Composition, x %	Absolute Viscosity, μ g/m-sec
Carbon Dioxide	16.45	0.057
Oxygen	1.89	0.061
Helium	79.43	0.054
Nitric Oxide	0.1	0.054
Methane	2.14	0.038

Viscosity of the mixture, $\mu_{\text{mix}} = \sum x\mu / \sum x = 5.43\text{E-}02 \text{ g/m-sec}$

Since the species are all in the gas phase, the viscosities of which are very negligible ($\mu_{\text{mix}} = 0.00543 \text{ g/m-s}$), we will ignore any wall effects, since it is also going to be negligible.

Reynolds Number, $Re = D v \rho_{\text{mix}} / \mu_{\text{mix}} = 20.6$

Since $Re < 2100$ for a flow of a fluid through a circular tube, we can conclude that the flow is laminar.

Mixing Chamber

The mixing chamber is just to prevent any surge inside the reactor and to create some back pressure in order to meter consistent and steady flow of various gases through various rotameters. The mixing chamber will be a stainless steel chamber. The chamber will be

designed to provide 10 seconds of flow rate buffer capacity i.e $1950 \text{ cc/min} \times (10/60) = 325 \text{ cc}$.

Assuming 5 cm diameter, the length is 10 cm.

TEST MATRIX

The research team at Rust College would like to develop data such as the extent of NO reduction with reburning fuels like (a) $\text{CH}_4 + \text{C}_2\text{H}_2$ (b) $\text{CH}_4 + \text{NH}_3$ during the first year. For each of these reburning fuels, the focus will be to find the operating point i.e what reburning stoichiometric ratio (SR2) is optimum out of SR2 range 0.75-1.0.

The following table lists the test matrix for the Year 1.

Experiment	Test conditions	Objective of the test	No. of runs
1. Reburning with CH_4 (Validity tests for the experimental facility)	a. Temp=1373 K Pressure=1 atm. Reburning Stoichiometric ratios (SR2): 0.75, 0.8, 0.85, 0.9, 0.95, 1.0.	To test if our experimental setup can achieve what is projected in computer runs & previous researchers' experiments.	SR2=0.75, 0.8, 0.85, 0.9, 0.95, 1.0. 6 runs
	b. Temp=1273 K Pressure=1 atm. SR2=±0.1 of best SR2 from 1a.	To test if lowering temperature has any impact.	3 runs (SR2 listed in test conditions)
	c. Same as 1b But Temp=1473 K	To test if increasing temperature has any impact.	3 runs (SR2 as in test conditions)

2.Reburning with $\text{CH}_4/\text{C}_2\text{H}_2$	a. Temp=1373 K or best operating temp point from 1b & 1c. Pressure= 1 atm. $\text{CH}_4=90\%$ and $\text{C}_2\text{H}_2=10\%$ SR2=0.75,0.8,0.85, 0.9,0.95,1.0.	To reduce NO to levels below what CH_4 can individually achieve.	6 runs
	b. Same as 2a but $\text{CH}_4=80\%$, $\text{C}_2\text{H}_2=20\%$ SR2= ± 0.1 of best SR2 from 2a.	To see if we can get better reduction than 2a.	3 runs
	c. Interpret from 2a & 2b as what will be best $\text{CH}_4=?$, $\text{C}_2\text{H}_2=?$. SR2= ± 0.1 of best SR2 from 2a.	To keep the utility cost of C_2H_2 down at the same time achieving the necessary NO reduction.	3 runs
3.Reburning with CH_4 & NH_3 .	a.Temp=1373 K or best operating point from 1b & 1c. Pressure= 1 atm. SR2=0.75,0.8,0.85,0.9,0.95,1.0. $\text{CH}_4=96\%$ $\text{NH}_3=4\%$	To reduce NO to levels below what CH_4 can individually achieve. To find if it will be better than CH_4 & C_2H_2 i.e 2a & 2b.	6 runs
	b.same as 2a but $\text{CH}_4=92\%$, $\text{NH}_3=8\%$ SR2= ± 0.1 of best SR2 from 3a.	To find if we can get better reduction.	3 runs
	C. Interpolate from 3a & 3b as what will be the best $\text{CH}_4=?$ $\text{NH}_3=?$ SR2= ± 0.1 of best SR2 from 3a.	To keep the utility cost of NH_3 down and at the same time achieving the necessary reduction.	3 runs

RATE OF GAS HEATING

Rate of gas heating calculations were performed using the radiative heat transfer to the gas mixture. However, another calculation was also performed based on pure convection since Dr. Ekman insisted that the model not include radiation to verify whether the gas actually gets heated to the desired temperature within the time of its residence. Both methods yielded favorable result. The details are presented below.

(a) Radiative heat transfer to the gas mixture

Heat gained by the gas mixture = Heat given by the radiative element (furnace walls)

$$mC_p dT = \sigma A \epsilon (T_w^4 - T^4) dt$$

On rearranging,

$$t = \frac{m}{\sigma A \epsilon} \int \frac{C_p dT}{T_w^4 - T^4}$$

where T_w is 1373 K and the integral limits are from 288 K (initial temperature of the gas mixture) to 1372 K (the temperature to which gas needs to be heated).

Also, mass of the mixture, m equals 2.26E-07 kg-mol (1 g/min divided by the M.W of the mixture, 11.4 times 0.1555 s/ 60 s times 1E-03 for kg/g conversion).

The Stefan-Boltzmann constant is 5.669E-08 W/m² /K⁴; emissivity of ceramic is 0.3 and

surface area, A is given by $\pi (.01885) (0.09125) \text{ m}^2$.

C_p of the gas mixture is $4.915 + 2.584E-03 T - 1.6E-06 T^2 + 3.92E-10 T^3 \text{ kcal/kg-mol/K}$

The integral now becomes:

$$\int \frac{C_p dT}{T_w^4 - T^4} = 4.915 \int \frac{dT}{T_w^4 - T^4} + 2.584E-03 \int \frac{T dT}{T_w^4 - T^4} - 1.6E-06 \int \frac{T^2 dT}{T_w^4 - T^4} + 3.92E-10 \int \frac{T^3 dT}{T_w^4 - T^4}$$

Upon integration this resolves to the following to which the integration limits (288 K to 1323 K or whichever T chosen) are to be applied.

$$\begin{aligned} & 4.915 * \frac{1}{4T_w^3} \left[\ln \frac{T_w + T}{T_w - T} + 2 \arctan \frac{T}{T_w} \right] \\ & + 2.584E-03 * \frac{1}{4T_w^2} \ln \frac{T_w^2 + T^2}{T_w^2 - T^2} \\ & - 1.6E-06 * \frac{1}{4T_w} \left[\ln \frac{T_w + T}{T_w - T} + 2 \arctan \frac{T}{T_w} \right] \\ & + 3.92E-10 * -\frac{1}{4} \ln [T_w^4 - T^4] \end{aligned}$$

Upon applying integration limits it is found that it takes 0.0477 seconds to heat the gas from 288 K to 1372 K which is very encouraging to the use of this particular reactor.

Please refer to the residence time calculation (in the heating zone of the reactor). It would take 0.1555 s for the gas at 1100 C to travel 9.125 cm moving at 59 cm/s. The time required

to heat the gas from 288 K to 1372 K is $0.0477 \text{ s} \ll 0.1555 \text{ s}$. So the gas will be at the appropriate temperature (1373 K) when it enters the reburning zone.

(b) Heat transfer by convection

Question: Without radiation into account, for a laminar flow, what length of the tube is needed such that the exit bulk temperature of the gas is 1323 K? Take inlet temperature of the gas mixture as 273 K. The wall temperature is 1373 K (furnace temperature).

Velocity of the gas mixture, $v = 0.5868 \text{ m/s}$

Diameter of the tube, $d = 0.01885 \text{ m}$

$$T_w = 1373 \text{ K}$$

The heat transfer balance is given by

$$q = h \pi d L \left(T_w - \frac{T_{b1} + T_{b2}}{2} \right) = \dot{m} c_p (T_{b2} - T_{b1})$$

Substituting temperatures and the diameter, this simplifies to:

$$31.09 h L = 1050 \dot{m} c_p$$

The quantities $\dot{m}$ and c_p need to be calculated based on mean temperature ($T_{\text{mean}} = 0.5(T_{b1} + T_{b2}) = 798 \text{ K}$).

The composition of the gaseous mixture at $SR2 = 0.9$ along with various properties at 800 K

is given below.

Gas	% comp.	Density kg/m ³	Viscosity kg/m-s	Specific heat J/kg-C	thermal conductivity W/m-C
CO ₂	16.45	0.671	3.39 E-05	1126	0.0560
O ₂	1.89	0.488	4.21 E-05	1054	0.0603
CH ₄	2.14	0.2446	2.40 E-05	3936	0.1046
He	79.43	0.061	3.88 E-05	5197	0.3070
NO	0.1 (Negligible influence on the mixture properties)				

Mixture Properties

0.07467 3.774E-05 4422.6 0.2567

mass flow rate, $\dot{m} = \rho (\pi d^2/4) v = .07467 * (\pi * .01885^2/4) * 0.5868 = 1.2228E-05 \text{ kg/s}$

The heat balance equation becomes upon substituting m and c_p :

$$h L = 1.8264 \text{ W/m-C}$$

Reynolds Number, $Re_d = \rho v d / \mu = .07467 * .5868 * .01885 / 3.774E-05 = 21.885$

This confirms the laminar flow assumption.

Prandtl Number $= \mu c_p / k = 0.65$

Peclet Number, $Pe = Re_d Pr = 14.225$

Hausen has developed the following formula for the Nusselt Number (N_d) for fully developed laminar flow with constant wall temperature scenario which is valid for all ranges of Peclet Number*(d/L).

$$N_d = 3.66 + \frac{0.0668 \text{ } Pe \frac{d}{L}}{1 + 0.04 \left(Pe \frac{d}{L} \right)^{\frac{2}{3}}}$$

Since heat transfer coefficient, h is dependent on the length, L, let us choose a particular

length, find h and calculate L based on the heat transfer balance equation, $h L = 1.8264$.
Reiterate the procedure until L converges to the actual length needed to heat the gas in the tube to the desired temperature.

Choose $L = 5 \text{ cm} = 0.05 \text{ m}$

$$Pe \, d/L = 14.225 * 0.01885 / 0.05 = 5.363$$

$$Nu_d = 3.979$$

$$h = Nu_d k / d = 3.979 * 0.2567 / 0.01885 = 54.19 \text{ W/m}^2\text{-C}$$

$$\text{Therefore } L = 1.8264 / h = 1.8264 / 54.19 = 0.0337 \text{ m} = 3.37 \text{ cm}$$

Reiterate:

$$Pe \, d/L = 7.9556 \quad Nu_d = 4.1184 \quad h = 56.08 \text{ W/m}^2\text{-C} \quad L = 0.0326 \text{ m} = 3.26 \text{ cm}$$

Reiterate:

$$Pe \, d/L = 8.2339 \quad Nu_d = 4.1329 \quad h = 56.28 \text{ W/m}^2\text{-C} \quad L = 0.03245 \text{ m} = 3.25 \text{ cm}$$

We thus find that the length of the tubular reactor needed to heat the gas entering at a velocity of 0.5868 m/s from 273 K to 1323 K, with wall temperature maintained at 1373 K is 3.25 cm.

Also, the time taken to achieve this heating is very small (0.0325 m divided by 0.5868 m/s):
0.0553 s.

ANSWERS TO OTHER QUESTIONS/CONCERNS

1. What type of NO_x analyzer is used? Gases other than NO_x should be analyzed so that a mass balance can be performed around the experimental apparatus.

The NOx analyzer that will be used is a chemiluminiscent NOx analyzer. The purchase order has been placed with California Analytical Instruments (Model 300 CLD). This measures 8 ranges of ppm from as low as 0-3 ppm range to as high as 0-30,000 ppm range.

The major gases exiting other than NO will be N_2 , CO_2 , CO, HCN, and NH_3 .

a. CO, CO_2 , CH_4 , O_2 can be analyzed by a Non-Destructive Infrared (NDIR) analyzer (Model 1200 Fisher 11-128-1).

b. NH_3 can be analyzed by specific ion electrodes (Orion Model 9512 BN) Range (1-1000 ppm).

c. HCN can be analyzed by Orion Model 94-06 BN along with a double junction reference electrode and an ionic strength adjuster. Range: 1-1000 ppm

Both b. and c. would require an output meter (simultaneous, dual output) No. 0720A3.

Unfortunately none of the above have been included in the budget for equipment. While we see the merit of performing mass balance around the apparatus, the budget constraints limit us from doing so. During the course of the project efforts will be made to obtain funds for this purpose.

2. Are catalytic effects probably negligible for PC firing?

Burch et al (1991) have reported that lignite, char, ash reduce NO from 1000 ppm to <50 ppm at pulverized coal combustion conditions. They have also reported that lignite ash contains a major portion of CaO (28.2%), Barium (6570 ppm) and Strontium (4900 ppm). They have suggested that these seem to be the most likely candidates for catalysts. The kind of reduction reported is not negligible and suggests that catalytic effects can be significant.

3. The use of helium to adjust the heating rate, due to its lower thermal capacity removes

from the study from realistic conditions.

Helium is used in this experiment for blending the gas (dilution) and to get the required flow rate of 1950 cc/min . The main reason helium is used is to minimize the heating time. Furthermore, helium similar to nitrogen is inert and hence will not remove the study from realistic conditions. As mentioned in our technical report the simulated flue gas will consist of helium base producing rapid heating and cooling.

Usually industries use 10% excess air to burn the coal completely. We assumed the same 10% excess O_2 and obtained the simulated flue gas composition: pure CO_2 - 327.67 cc/min, pure O_2 - 37.62 cc/min and NO - 1.95 cc/min. We assumed the primary zone fuel to be Pittsburgh coal.

Upon our calculation, energy required to heat 1 kg-mol of gas mixture from 25 C to 1100 C with Helium as the diluent gas is 6585 kcal where as the heat required to do the same with N_2 as diluent gas is 8611 kcal. So, in order to heat the gas mixture with N_2 as diluent, we need 30% more heat energy and hence more heating time. Also, Helium has been used as diluent gas by the following researchers, to cite a few.

- i. Levy, J.M. , L.K. Chan, A.F.Sarofim and J.M.Beer (1981) and
- ii. Singoredjo, L., F. Kapteijn, J. A. Moulijn and H.P. Boehm (1993).

4. The modeling analysis and experimental plan stop with the upper fuel injection. What happens to the reaction products when the remaining combustion air is added? Making significant HCN does not preclude reaction of the HCN to NO in the presence of oxygen.

Our study concerns with reducing NO in the reburning zone. We have not proposed to study by modeling or experimental analysis what will happen in the burnout zone. This would

require a lot more labor, capital than what is allotted for this study. However, we will be glad to do the computer modeling study in the burnout zone. In the burnout zone we will use excess O_2 -10% and $T=1200K$ (Kilpinen et al, 1992) or 1373K and watch for HCN reduction and (N_2O formation). Song et al (1982) have published the decomposition of HCN as a function of furnace temperature. It is shown that HCN is highly unstable at high temperatures and its recovery is reported to be 30% for a residence time of 1s at 1100 C. Hence, the high HCN observed at lower stoichiometries could be overcome by the burnout zone. We will be glad to do a computer modeling study where we will just input the flue gas from reburning zone & we will then introduce O_2 & observe HCN reduction.

To computationally test HCN conversion to N_2 or NO_x after reburning:

- a. We will input HCN (1000 ppm) and Oxygen at $SR3=1.25$ (optimum as per S.L. Chen et al) into the burnout zone and find NO formation. The temperature of the burnout zone will be 1500F (found to be optimum by S.L. Chen et al). The residence time will be 0.6 s for burnout zone; this residence time is recommended by Angelos Kokkinos (1992).
- b. We will alternatively input the reburning products from the reburning zone at $SR2=0.9$ and input this into burnout zone with $SR3=1.25$.

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