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REDUCTION OF NOX AND CO TO BELOW 2PPM IN A DIFFUSION FLAME

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ABSTRACT

The so-called "sudden death reaction" theory, for a diffusion flame, assumes that the fuel and oxidizer diffuse toward a stoichiometric concentration surface, and then suddenly disappear, due to their combustion which produces water and carbon dioxide. The presence of NOx and CO in the combustion products cannot be explained by the "sudden death" theory. NOx, due to its high activation energy may not be formed prior to the formation of H_2O and CO_2. NOx is created when both oxygen and nitrogen are present in a high temperature volume; after all the combustible species are consumed. Appearance of CO indicates a lack of oxygen or a low gaseous temperature. Traditionally, when steam is injected into the combustion air, its high heat capacity reduces the flame temperature, which then reduces NOx formation, and this is usually accompanied by high CO formation. This phenomenon is caused by the dilution of oxygen as a quenching effect.

This paper describes a novel approach that reverses the traditional wisdom of using steam to control NOx and CO formation, by accelerating the combustion process. This new approach begins with (1) shrinking the flame envelope, (2) enhancing the oxygen diffusion rate, and (3) suppressing the nitrogen concentration diffusion rate. Test results showed that (1) a high temperature volume could form NOx after the combustion of fuel is reduced to a minimum, and (2) that a very high fuel jet momentum increases the oxygen diffusion rate, thus reducing the flame envelope. Also due to the inward movement of the flame envelope, the residential time for NOx formation is also reduced and with the presence of a diluent, the nitrogen penetration rate into the flame is controlled. When all three phenomena are working together, total NOx was reduced downward to below 2 ppm without losing flame stability. Since this process generates enhanced oxygen diffusion, CO has always been seen to be below 2ppm, which indicates extremely high combustion efficiency.

The above theory was first simulated by numerical methods using a 3-step reaction for nitrogen and oxygen, and was further expanded to a 28-step chemical kinetic model. The simulation used gas turbine compressor discharge temperatures to produce real adiabatic flame temperatures. Atmospheric tests of real full-scale gas turbine combustors were used with appropriate air temperatures, to simulate adiabatic flame temperatures. Below 2ppm NOx and CO were consistently obtained, independent of turbine types. Actual turbine tests on GE 6B and W501D5A turbines consistently indicated pressure dependent exponents of 0.1.

INTRODUCTION

Flames can be classified into as diffusion and premixed flames. Most combustion that occurs in gas turbines is based on the diffusion flame principle. Fluid mechanical coupling to the diffusion process has dominated combustor design principles for decades. The design and development of low-emission industrial gas combustors is very challenging. From a realistic point of view, presence of NOx and CO in the combustion products, illustrates the need to perform an in-depth study of the diffusion flame. Previous techniques to modify combustors to control emissions have focused on lowering the peak flame temperatures. This paper reports on a novel approach that reverses the traditional wisdom of using steam to cool off a diffusion flame. First, the theory is explained in detail, and then numerical calculations were performed to demonstrate how this theory works. Then, this theory was proven experimentally. The theory was atmospherically tested on full-scale gas turbine combustors with an appropriate air supply temperature to simulate adiabatic flame temperatures. Below 2ppm NOx and

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CO were consistently obtained independent of turbine type. Actual turbine tests on a GE 6B engine used for providing commercially available power indicated pressure dependent exponents of 0.1. Therefore, the NOx levels were found to reduce down to below 2 ppm without losing flame stability. Since this process uses enhanced oxygen supply, CO was found always to be below 2ppm, which means that there was extremely high combustion efficiency. Unburned hydrocarbons (VOC) have always been found to be undetectable.

NOMENCLATURE

t	Time
ρ	Density
$\vec{V}$	Velocity
P	Pressure
m_{fi}	Mass Fraction of Species Component i
ν	Kinematic Viscosity
D	Diffusivity
λ	Thermal Conductivity
Cp	Specific Heat
T	Temperature
m_{fi}'''	Chemical Volume Reaction Rate
H'''	Volume Heat Generation
u	x Direction Flow Velocity Component
v	y Direction Flow Velocity Component
K	Jet strength
η	Similarity Variable
ε_o	Virtual Kinematic Viscosity
b	Jet Width
U	Jet Velocity

THEORY DESCRIPTION

Classical diffusion flame theory is based on the so-called "sudden death" reaction theory. This theory is illustrated in Figure 1b. The top figure shows the diffusion profiles of a fuel and oxidizer before combustion. The location where the flame surface would occur is at the stoichiometric ratio (kg oxidizer over kg fuel). In this illustration, the value of this ratio is one. The bottom figure shows the diffusion profiles during combustion (Glassman, 1996). As shown, the theory assumes that the fuel and oxidizer diffuse towards a stoichiometric concentration surface. According to this theory, both fuel and oxidizer suddenly disappear, by combustion at an infinitely thin flame surface, which produces combustion products such as water and carbon dioxide. This theory assumes there are infinitely fast chemical reaction rates, so that the burning speed is controlled by the molecular and turbulent diffusion rates of the fuel and oxidizer.

Figure 1a shows the effect of the traditional way of using steam for emissions control, by mixing steam in the air. This technique has been used as a means to lower the peak flame temperature, thus minimizing the thermal NOx formation. However, by mixing the steam in the air, the concentration of oxygen is reduced. Since, the concentration of oxygen is reduced, the flame surface moves outward. Also, the quenching of the flame can lead to increased CO formation due to the lack of oxygen.

Our new approach for emissions control (Cheng, 2002) is shown in Figure 1c and involves a homogeneous mixing of the fuel with a diluent such as steam. By mixing the fuel with the diluent, we lower the concentration of fuel, and the result is that the stoichiometric surface is brought inward. In this process, we now have an excess of oxidizer; therefore the CO and unburned hydrocarbons are reduced.

Figures 1 and 2 also show the concentration profile of Nitrogen in the flame. Notice that oxygen cannot penetrate the flame front since it reacts with the fuel at the stoichiometric surface. There is also a presence of Nitrogen within the flame front. The effect of our new approach for emissions control is illustrated in Figure 1c and by the results of our numerical simulation in Figure 2. The presence of steam in the high temperature region, where NOx formation is at a maximum, reduces the presence of Nitrogen. Figure 2 shows that in the region where maximum NOx is formed, the mass fraction of nitrogen is reduced, thus the NOx formation is also reduced.

Figure 3 (Lewis and Von Elbe, 1951) shows the flame length for laminar and turbulent diffusion flames as a function of fuel jet velocity. In a laminar diffusion flame, the flame height increases with fuel jet velocity. The molecular diffusion rate causes the flame surface area to be proportional to the jet velocity. In the turbulent flame, the high fuel jet velocity creates turbulent mixing which increases the molecular diffusion rate. The region of constant flame height in the fully developed region indicates a similarity flow condition where the turbulent mixing profile becomes independent of velocity. In this region, increases in the fuel jet velocity do not affect the flame height.

The governing equations (Schliting, 1968) for turbulent diffusion flames are given below.

Continuity: $$\partial\rho/\partial t + \nabla\bullet\rho\vec{V} = 0 \tag{1}$$

Momentum: $$\partial\vec{V}/\partial t + \vec{V}\bullet\nabla\vec{V} = -\nabla P + \nu\nabla^2\vec{V} \tag{2}$$

Mass Transfer: $$\rho(\partial m_{fi}/\partial t + \vec{V}\bullet\nabla m_{fi}) = \rho D\nabla^2 m_{fi} + m_{fi}''' \tag{3}$$
$i = 1,2,3\ldots n$

Heat Transfer: $$\rho Cp(\partial T/\partial t + \vec{V}\bullet\nabla T) = \lambda\nabla^2 T + H''' \tag{4}$$

In the above equations: ρ is the density, $\vec{V}$ is the velocity vector (which is equal to the sum of the time averaged velocity and velocity of fluctuation), t is time, P is pressure, ν is the kinematic viscosity, m_f is the mass fraction of the species

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component, D is the diffusivity, m_f''' is the chemical volume reaction rate, Cp is the specific heat, λ is the thermal conductivity, T is the temperature, and H is the heat generated due to combustion. The transport properties such as ν, D, and λ (kinematic viscosity, diffusivity, and thermal conductivity) are equal to the sum of the molecular value and the turbulent mixing value. When the equations are put into their non-dimensional form, the Reynolds number, Peclet number, Schmidt number, and Prandtl number can appear. At high turbulent conditions, the turbulent transport theory states that the Schmidt number, Lewis number, and Prandtl number each approach 1. Thus, the velocity, concentration, and temperature profiles become similar if there is no combustion.

The viscous flow forms of these turbulent flow equations are solved to obtain the similarity solution for the flow of jets (Schliting, 1968). The similarity solution requires that there is no length scale for the problem. The similarity variable (η) reduces the partial differential equation into an ordinary differential equation. The solution gives the following equations for both velocity components:

$$u = (3/8\pi) * (K/\varepsilon_o x) * (1/(1+1/4\ \eta^2)^2) \tag{5}$$

$$v = (3/16\pi)^{0.5} *(K)^{0.5}/x * ((\eta - 0.25\eta^3)/(1+1/4\eta^2)^2) \tag{6}$$

where $\eta = (3/16\pi)^{0.5} * (K^{0.5}/\varepsilon_o) * y/x$ is the similarity variable, $K = 2\pi\int u^2 y dy$ is the jet strength, and $\varepsilon_o = 0.0256 b_{1/2} U$ is the virtual kinematic viscosity (b is the jet width and U is the jet velocity). It is important to note in this solution, that the variable x still appears. Thus, the length scale still remains an important parameter in the solution.

Figure 4 illustrates the streamline pattern (Schliting, 1968) for an axisymmetric fuel jet in this similarity flow condition regime. At high Reynolds number (high momentum conditions), the jet will remain narrow and grow slowly. Also as the jet momentum increases, the radial changes in the velocity will also increase. Therefore, increasing the fuel jet momentum by homogeneous mixing with a diluent will increase the oxygen diffusion rate into the flame.

Research work in our laboratory (both theoretical and experimental) with known chemical kinetics, has made it obvious that NOx due to its high activation energy may not be formed at the flame front since it blocks oxygen penetration, rather it was created when both oxygen and nitrogen can be present in a small volume of high temperature after all combustible species are consumed as illustrated in Figure 5. Presence of CO is an indication of lack of oxidizer or lower gaseous mixture temperatures. When attempts are made to inject steam, water, or to use cooled catalytic combustion, NOx will be reduced but usually is accompanied by high CO production due to dilution of oxygen concentration thus it quenches the combustion process. In contrast we enhance the oxygen concentration. Figure 5 shows the difference with our new technology (Cheng, 2002) to lower emissions compared to the traditional steam injection method. Our approach begins with (1) shrinking the diffusion flame envelope, (2) enhancing the oxygen diffusion rate, and (3) suppressing the nitrogen concentration diffusion rate. These result in (1) the highest temperature zone volume after the combustion of fuel is reduced to a minimum, and (2) a very high fuel jet momentum which reduces the flame envelope volume. This is simultaneously done by homogeneously mixing the fuel with the diluent, so that it brings the stoichiometric concentration surface of fuel and oxygen inward. The higher volume flow requires a higher jet momentum issuing from the fuel nozzle, which further enhances the convective component of oxygen to the molecular diffusion rate. Due to inward movement of the flame envelope and several factors of a higher convective component after combustion, the residential time for oxygen and nitrogen to be simultaneously present in the small hot volume at the flame tip is reduced by orders of magnitude. Our approach involves a fuel side dilution (which has been explained earlier) and that the flame front will move toward the fuel side. As a result the temperature gradient increases and the diffusion rate and concentration gradient increases, which accelerates the combustion rate.

NUMERICAL SIMULATION OF THEORY

The above theory was simulated with numerical model techniques. The simulation uses gas turbine compressor discharge temperatures, to produce real adiabatic flame temperatures, for gas turbine applications. A commercially available computer program (Star-CD) was used for the simulations. This program uses computational fluid dynamic (CFD) modeling to determine the air and fuel interaction and how combustion takes place in the combustion chamber. This code has been used to model the high-pressure turbulent compressible flow that occurs in the combustion of methane in air for the Westinghouse 501D5A combustion geometry. The industry standard k-ε turbulence model was used in the simulations. The numerical technique uses a 3-step extended thermal reaction model for nitrogen and oxygen (Flowers, 1974). The results from this mechanism have been shown to agree well with experimental data of NOx formation in a flame tube (Duer, et.al. 2000).

The fuel nozzle hardware used in W501D5A consists of 12 holes near the center of the nozzle used for traditional steam injection. Surrounding these steam holes are 12 holes used for the fuel injection. The high-pressure turbulent combustion for one fuel nozzle hole of the W501D5A combustion system was simulated. Fuel nozzle hole diameter is 5.08 mm. Combustion chamber diameter is 304.8 mm and length is 152.4 mm long. The fuel and airflow rates were set to give a total ignition temperature of 1505 K, a Pressure Ratio of 14:1, and compressor discharge temperature was set to 644 K. Three cases were studied for this configuration. The first case examines the combustion of methane in air at engine operating conditions. The second and third cases examine the effect of the

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approach of mixing the fuel with steam for two different ratios without changing the nozzle diameter. The first ratio involved adding an equal mass flow of steam with methane. The fuel jet momentum doubles for this ratio. The second ratio was a 3:1 steam to methane mass flow ratio (ratio of kg steam to kg methane). The fuel jet momentum for this ratio is now four times the original momentum. The next set of figures show the results for these three cases.

Figure 6 shows the fuel mass flow rate profiles for these three cases. The original engine operating case is on the left. It takes more than 200 fuel nozzle diameters downstream before the tip of the flame is reached. The flame length is smaller due to the accelerated combustion that occurs because of the increased fuel jet momentum. Our results show that with steam to fuel ratios above 3:1, the flame length does not change, i.e. similarity flow characteristics. Figure 7 shows the temperature profiles for these three cases. The results show that the maximum temperature region occurs after the fuel is consumed. Also as shown in the temperature profiles, the steam and fuel mixture lowers the maximum temperature by 95 K for the 1:1 steam to fuel ratio case and by 300 K for the 3:1 steam to fuel ratio case due to the high heat capacity of the steam. Figure 8 shows the mass fraction profiles of carbon monoxide. The results show that the production of CO stays approximately the same between the three cases. The maximum values occur at the edges of the flame. The amount of CO production does not increase with the homogeneous mixture of steam and fuel. Figure 9 shows the mass fraction NOx profiles for these three cases. The region of maximum NOx formation occurs at the tip of the flame, after the fuels have been consumed. The maximum NOx mass fraction is reduced by a factor of three when the momentum is double, and then by two orders of magnitude at a steam to fuel ratio of 3:1. The benefit of homogeneously mixing of steam with the fuel is that the steam does not oppose the convective flux of the combustion products.

The computer results illustrate how this approach of homogenously mixing steam with fuel to lower emissions works. The results from the simulations show: a) maximum values of NOx occur after the fuel is consumed, b) the homogeneous mixture of steam and fuel results in a higher momentum jet which enhances the oxygen diffusion to the flame, c) the steam and fuel mixture accelerates the combustion which results in a higher temperature gradient in the flame, and d) the presence of steam reduces the nitrogen concentration and thus reduces NOx emissions and due to the presence of more oxygen the CO stays at low levels. The high levels of oxygen are also the reason for no unburned hydrocarbons.

ATMOSPHERIC AND ACTUAL ENGINE RESULTS

Several atmospheric tests with this methodology have been performed with our test facility (Wang, 2002). The test facility enables full-size combustion testing of single combustor cans, transition piece, and fuel nozzle hardware used in commercial engines at atmospheric pressures with preheated air at the combustion discharge temperature. Several combustors from different commercially available engines have been tested with this new low NOx emissions methodology (Wang, 2002).

Several studies have been done recently in the area of test result scaling and pressure effect on NO_x and CO emissions (Bhargava 2000 and Riccio 2001). In order to determine the effect of pressure on emissions, atmospheric and actual engine tests have been performed. Since the combustion air in our atmospheric tests (Wang, 2002) are run at the compressor discharge temperature (CDT), the effect of adiabatic flame temperature on NO_x formation can be neglected. In general, the effect of pressure on NO_x formation can be expressed in the form of,

$$\frac{(NO_x)_1}{(NO_x)_0} = \left(\frac{P_1}{P_0}\right)^n \tag{7}$$

Where, P is the pressure of the combustion system, exponent n is the pressure dependence factor for NO_x formation, subscript “1” denotes an elevated pressure condition, and subscript “0” denotes an atmospheric condition.

Recent atmospheric test and actual commercially running engine results have been obtained for the GE Frame 6B Gas Turbine Engine. The actual engine data came from a GE PG6531B combustion turbine with Mark IV controls, Struthers-Wells HRSG and Coen duct burner located at the cogeneration facility in El Segundo, CA. An EPA certified laboratory (Fukuda, 2002) took the data, which was corrected, to a 15% oxygen level. This data in Table 1 shows the pressure dependence factor for various steam-to-fuel ratios. The results show that without a diluent in the fuel, the pressure dependence factor is quite high at 0.287. Then the pressure dependence factor drops exponentially to around 0.1 at high steam-to-fuel ratios. Also, the benefits of diluting the fuel with steam are shown in the NOx measurements. The engine test results show that at a steam to fuel ratio of 1.2 the NOx is lowered by 79% (decrease by a factor of 4.8). Similar results were obtained with the W501D5 engine (Sewell, 2001). The results from that test resulted in a Best Available Control Technology award designation for 2002 from the Texas Natural Resource Conservation Commission.

The results in Table 1 show that when the compressor discharge temperature is used in both the atmospheric and actual engine tests there is very low or no pressure dependence. This can be justified since the mass fraction of chemical species is similar and the turbulent transport properties are the same, therefore, the pressure dependence should be minimal.

Finally, results with the GE Frame 6B with our atmospheric test facility show how low NOx and CO values can be obtained with this new approach. Figure 10 shows the results of NOx and CO levels for different steam to fuel ratios for this hardware. The results show that NOx around 2 ppm and low CO levels are maintained when high steam-to-fuel ratios are employed. The flame was observed to be stable up to a steam to fuel ratio of 4 to 1. There were no excessive noise or pressure fluctuations

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observed. These results have been obtained as well for the other engines that have been tested.

CONCLUSIONS

This paper has presented a new approach to meet all the emission control rules if there is steam available such as in a cogeneration system or advanced Cheng cycle. The theory has been explained and demonstrated by computer simulations and has been proven by atmospheric experiments and actual commercial engine operations. Both NOx and CO were controlled to single digits without VOC. No ammonia is needed. This simple method can obtain the same levels of emissions such as DLE, DLN, and XONON with extremely throttable flame stability with long parts life due to its lower flame temperatures. This method increases the power output and lowers the heat rate when compared to all of the other methods. The investment can be justified by the amount of power increase alone as a payback, with NOx reduction as an added benefit.

ACKNOWLEDGMENTS

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Table 1 Comparison of GE 6B Atmospheric Test Results and the Actual Emission Measurement of a 6B Gas Turbine Engine Using the Steam/Fuel Homogeneous Low NOx Emission System (Cheng, 2002) with a Pressure Ratio of 11.

Steam/Fuel Ratio	Atmospheric Test NO_x Measurement Ppmvd, 15%O_2	Actual 6B Engine NO_x Measurement Ppmvd, 15%O_2	Calculated Pressure Dependence Factor, n (PR = 11)
0	63.34	126.1	0.287
1.2	18.74	26	0.137
1.3	17.11	24.1	0.143
1.4	15.50	19.8	0.102
1.5	13.94	17.7	0.100

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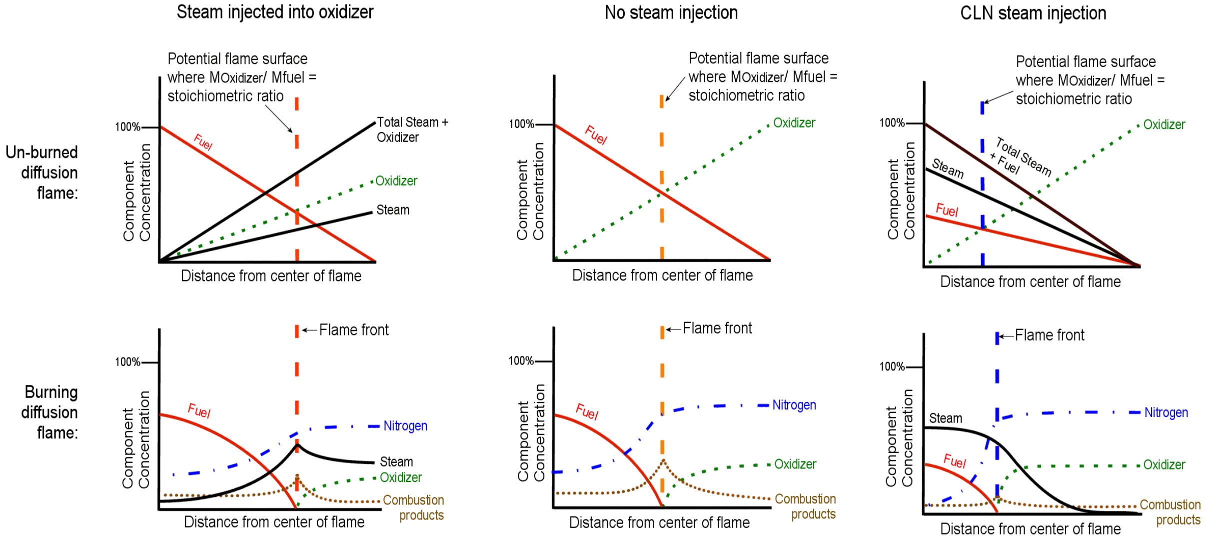

a b c

Figure 1. Illustration of diffusion flame concentration profiles diagrams for 3 cases. a) Component concentration profiles for before burning and after burning for steam injected into oxidizer. Note that the dilution of the oxidizer can cause the flame surface to move outward. b) Component concentration profiles for regular flame (no steam injection). c) Component concentration profiles for case of steam homogeneously mixed with fuel. Note that the dilution of the fuel causes the flame front surface to move inwards. Also note that nitrogen penetrates into the flame front while the oxidizer does not.

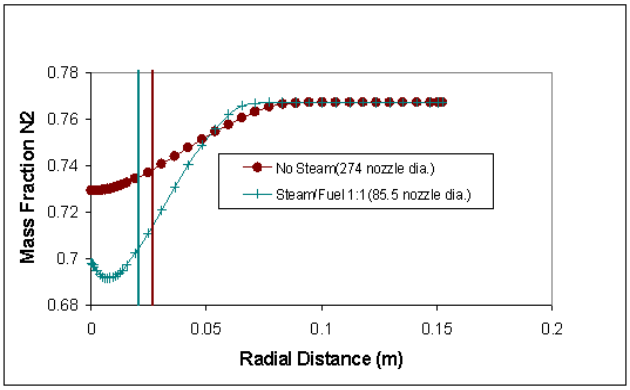

Figure 2. Numerical simulation results illustrating the nitrogen blocking effect at the mid-plane of the volume where maximum NOx is formed in the high temperature region. The lower concentration of nitrogen in this region yields less NOx formation. The solid vertical lines indicate frame front location (brown without steam, blue with steam). For steam mixed with fuel, the flame front moves inwards.

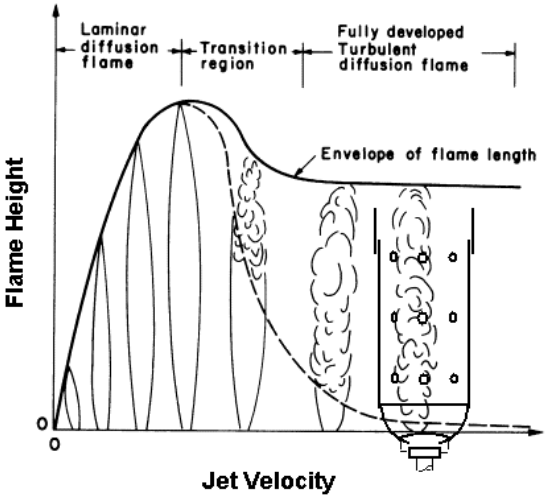

Figure 3. Flame height as a function of the fuel jet velocity. At high fuel jet velocities, the flame length becomes constant indicating a region of similarity flow.

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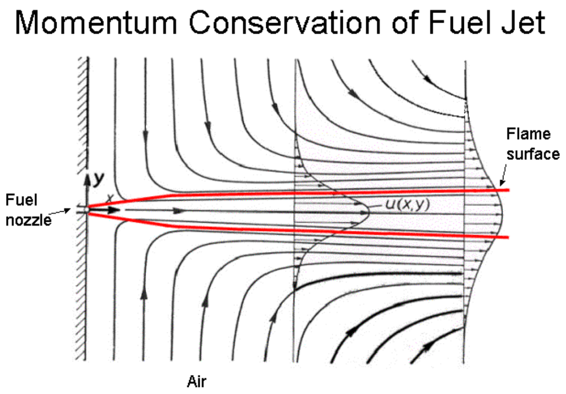

Figure 4. Similarity flow profiles for a circular turbulent jet showing the growth of the flame surface.

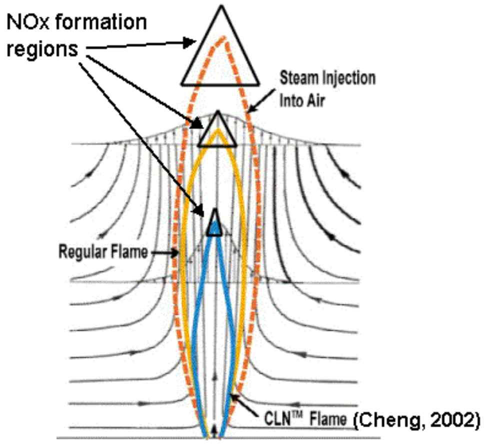

Figure 5. Reduction of the NOx formation region by homogeneously mixing fuel with a diluent (Cheng, 2002)

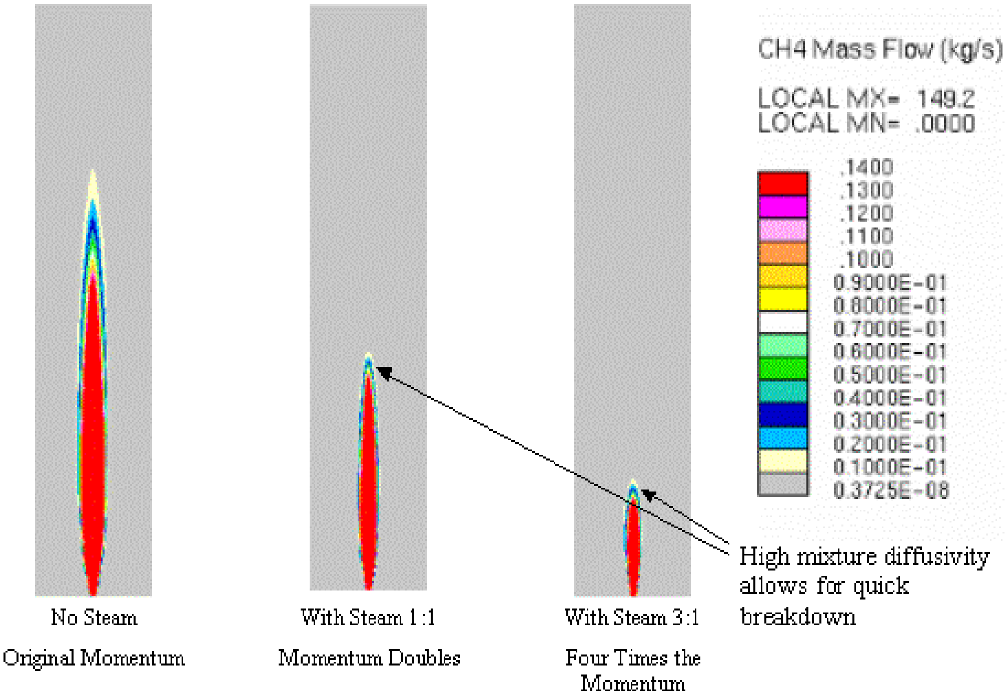

Figure 6. Numerical simulation results showing the effect of mixing steam with fuel on the variation of mass flow of methane. The steam to fuel ratios are expressed in terms of mass. At higher steam to fuel ratios (higher fuel and steam jet momentum), the fuel is quickly consumed indicating the combustion is accelerated.

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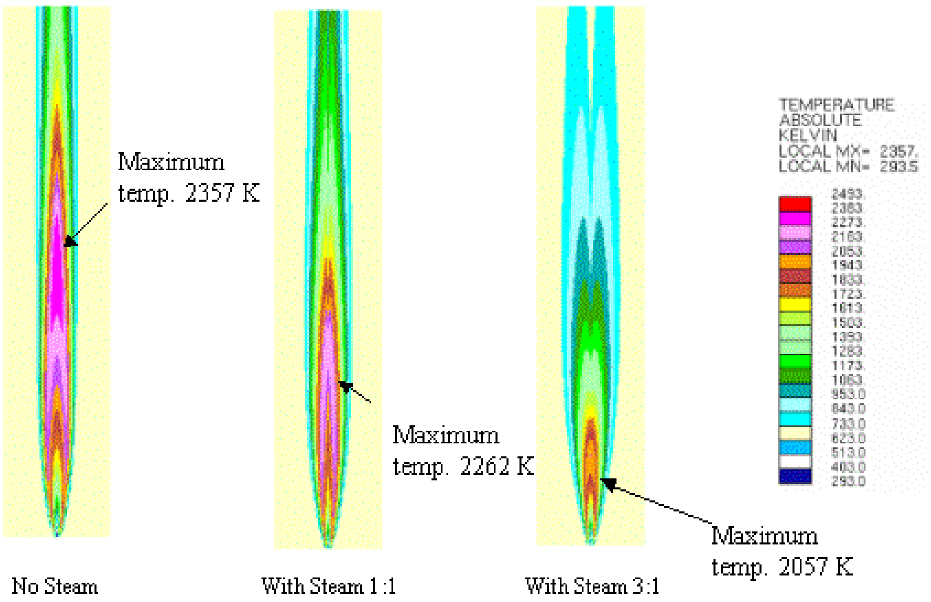

Maximum temp. 2357 K

Maximum temp. 2262 K

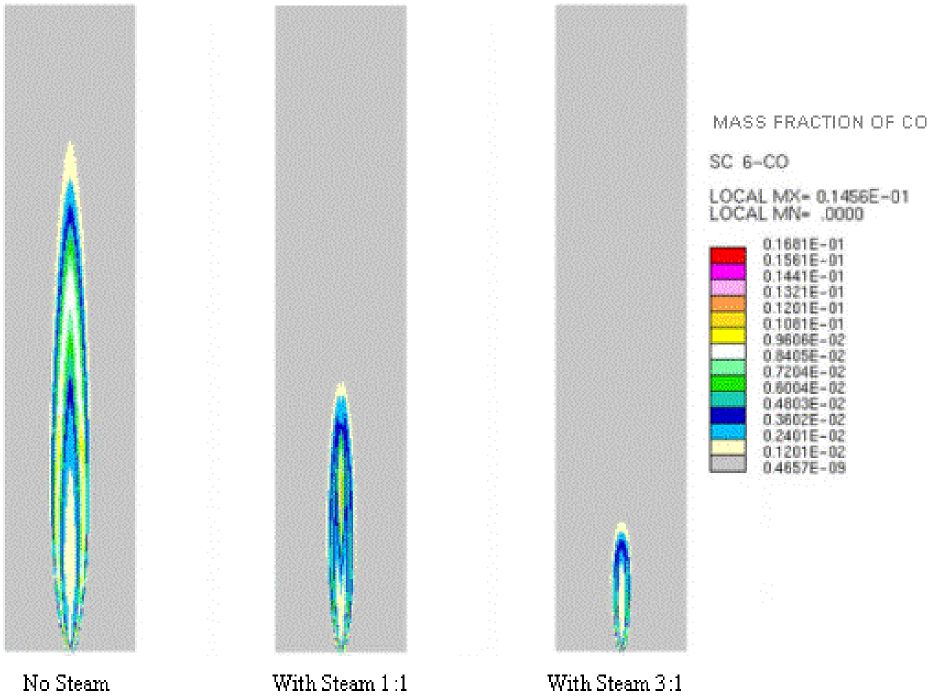
Maximum temp. 2057 K

No Steam

With Steam 1:1

With Steam 3:1

Figure 7. Simulation Results showing the effect of mixing steam with fuel on temperature profiles. The CFD results show that the temperature is decreased due to the high heat capacity of the steam, especially at high steam to fuel ratios.


No Steam

With Steam 1:1

With Steam 3:1

Figure 8. Simulation results show the effect of mixing steam with fuel on CO formation. The CO is expressed in terms of mass fraction. The results indicate no increase in CO formation is noticed by mixing steam with fuel.

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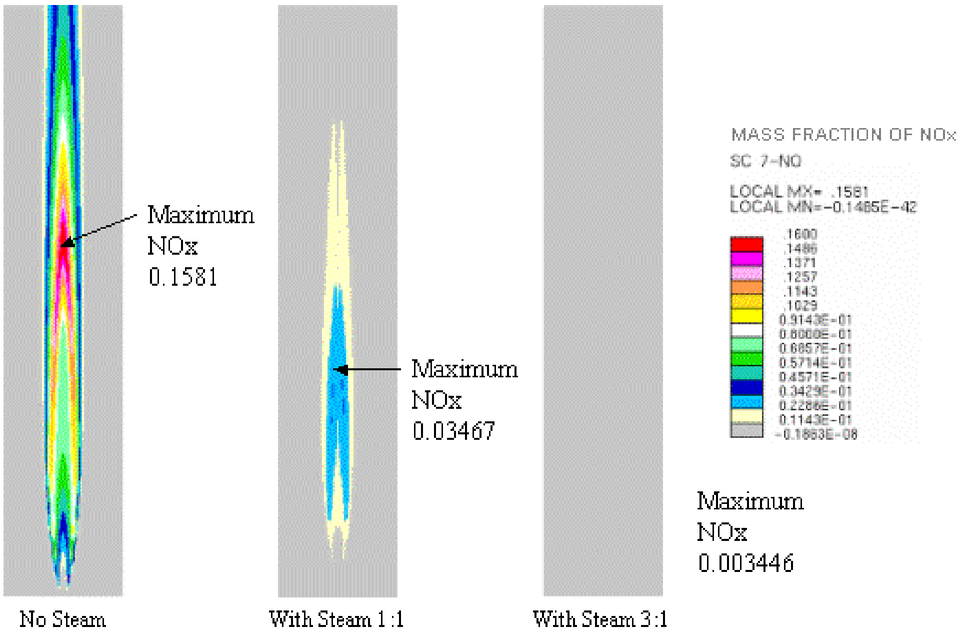

Figure 9. Simulation results show reduction of NOx formation by mixing steam with fuel. The NOx values and maximum NO values listed for each case are in terms of mass fraction.

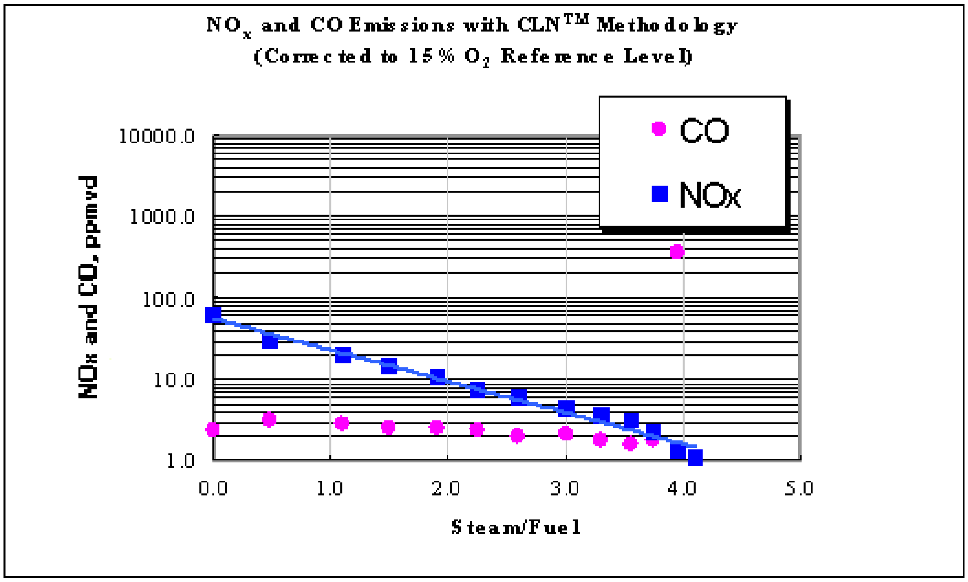

Figure 10. Experimental atmospheric test facility results at atmospheric pressure with the GE Frame 6B combustion liner demonstrating low NOx and CO values are consistently obtainable by mixing fuel with steam. At steam-to-fuel rations beyond 3.5 and before 4, the NOx is around 2 ppmvd and the CO remains at single digits.

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