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Published Citation

Smith, L. L., H. Karim, S. Etemad, and W. C. Pfefferle. "Catalytic combustion of gasified coal for low-emissions gas turbines." In 22nd Annual International Pittsburgh Coal Conference 2005.

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Catalytic Combustion of Gasified Coal for Low-Emissions Gas Turbines

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ABSTRACT

In response to the Department of Energy's (DOE) goals of developing low emission, coal-based power systems, Precision Combustion, Inc. is developing a low, single digit ppm NOx emissions system for high firing temperature IGCC systems. The present paper presents emissions data for syngas and alternate fuels tested successfully. Subscale testing results at 10 atmospheres include NOx emissions meeting DOE's target of 0.01 lbs/MMBtu (3 ppm at 15% O₂) and were below this value under some operating conditions during parametric testing.

INTRODUCTION

A new combustion method dubbed Rich-Catalytic Lean-burn (RCL[®]) was developed at Precision Combustion, Inc. (PCI) initially for operation on natural gas and has been successfully tested at full scale and in-engine at Solar Turbines [1]. The basic concept of this system is to catalytically react a portion of the fuel upstream of the combustor, thus preheating (and vitiating) the fuel/air mixture entering the combustor. The process of catalytic reaction improves combustion stability, especially at low flame temperatures. NOx emissions are improved by operating at lower allowable (stable) flame temperatures. Turndown to low engine power can be improved by operating at still lower flame temperatures without excessive emissions of CO or unburned hydrocarbons.

PCI, with DOE and gas turbine manufacturer support, initially developed this advanced catalytic combustor technology to offer ultra-low emissions through clean and efficient catalytic combustion for natural-gas-fired gas turbines. Originally developed under the U.S. Department of Energy's Small Business Innovation Research (SBIR) program, the technology offers simultaneous improvements in emissions, efficiency, fuel flexibility and component life. Currently, RCL is moving toward natural-gas-fired gas turbine field trial. The technology has retrofit potential and demonstrated successful operation with multiple fuels. Combustor module tests under large frame gas turbine conditions, fired with natural gas, have demonstrated the robustness of the technology providing stable combustion with NOx emissions as low as 2 ppm and low combustion dynamics. The RCL technology offers these benefits in a package sufficiently compact to potentially fit into existing frame machine combustor volumes.

Figure 1 depicts a schematic of the RCL combustion system.

As shown, the combustion air stream is split into two parts upstream of the catalyst bed. One part is mixed with all of the fuel and contacted with the catalyst, while the second part is used to backside cool the catalyst. At the exit of the catalytic reactor, the catalyzed fuel/air stream and the cooling air stream mix and burn to completion to provide the final burner outlet temperature. As pictured, combustion (fuel oxidation) occurs in two stages: a fuel-rich catalyst stage and an overall fuel-lean gas-phase combustion stage.

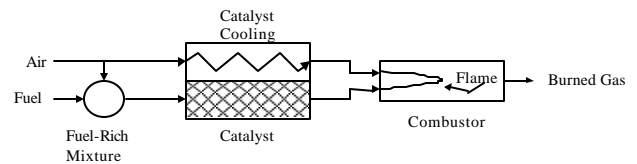


Figure 1. Schematic of fuel-rich catalytic combustion system, showing two-stage combustion process.

Currently, NOx emissions from conventional coal-fired power plants vary widely, from about 0.4 to 2.0 lbs/MMBtu (> 100-500 ppm) depending on burner type. Low NOx coal burners can reduce these emissions by about half, but ultra-low NOx emissions, to compete with natural-gas-fired turbines, requires alternative combustion means or after treatment.

One promising approach is coal gasification followed by combustion of the resulting syngas within a gas turbine engine. Integrated Gasification Combined Cycle (IGCC) power plants have been proven to achieve high efficiency with low emissions, including NOx emission guarantees of less than 25 ppmv at 15% O₂ (0.1 lbs/MMBtu) [2]. However, further reduction in NOx emissions by dilution of the fuel with inert gases faces barriers in terms of flame stability and impact on overall cycle efficiency. In fact, for highly diluted or low-Btu fuels such as nitrogen- or steam-diluted syngas, combustion may be unsustainable even at the highest possible flame temperatures unless flame stability is augmented by catalytic pre-reaction of a portion of the fuel.

The RCL combustion system is especially well suited for syngas fuels since it is designed to operate robustly and with constant performance using a wide range of fuels. In this paper we report test results using RCL technology for burning syngas for high firing F-class machines. In addition, test results for blast furnace gases and refinery fuels are also presented and discussed.

Submitted to the 22nd Annual International Pittsburgh Coal Conference, September 12 - 15, 2005.

HARDWARE CONFIGURATION AND OPERATING CONDITIONS

A sub-scale catalytic reactor for high-pressure testing with syngas fuel was fabricated at PCI, and is shown prior to final assembly in the photograph in Figure 2. The reactor housing is the long piece shown and flow is from the top-right to bottom-left of the photograph. During assembly, an injector for syngas fuel is fitted at the upstream end of the reactor where fuel and air mix to provide a fuel-rich fuel/air mixture to the catalyst. The large flange-like piece shown in the photograph contains the fuel plenum and the syngas fuel is delivered through the needle-like injectors shown. The reactor is fitted with a variety of instruments including thermocouples to measure catalyst and housing temperatures, flush-static pressure ports to measure reactor pressures, and gas sample extraction ports to measure gas composition entering and exiting the reactor. These instrumentation lines are coiled and visible in the photograph.

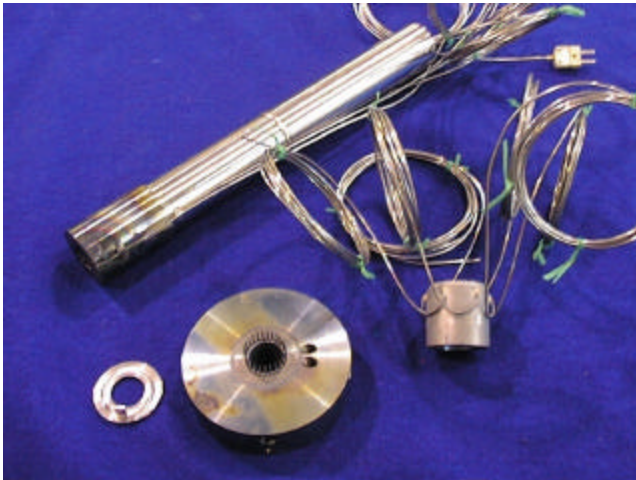


Figure 2. Photograph of sub-scale catalytic reactor for syngas combustion.

For high-pressure testing, the catalytic reactor of Figure 2 is inserted into the combustion test rig shown in Figure 3. Again, flow is from right to left, and the reactor is inserted at the right hand side of Figure 3. Two independently controllable air supplies are provided (both heated and at high pressure); the larger air supply (entering from the right in Figure 3) provides catalyst cooling air, which becomes primary zone combustion air in the gas-phase combustor, and the smaller air supply (entering from the vertical pipe at the top-right of Figure 3) provides air to the fuel-rich fuel/air mixture. For operation with syngas fuel, two heaters are also provided (but not shown in this photograph); one heater heats N_2 diluent just before it is mixed with fuel, and the second heater heats all other fuel components and CO_2 .

Downstream of the reactor, the catalytically reacted gases and the catalyst cooling air burn in the high-pressure ‘combustor burnout section’ labeled in Figure 3. Here, gas-phase combustion leaves the 0.6-inch reactor and completes the burnout of fuel within a 2-inch inside-diameter ceramic combustor liner, giving about a 10:1 dump ratio on an area basis. The combustor burnout section is instrumented with 6 S-type thermocouples to measure flame temperatures axially along the combustor liner at 3-inch increments, and 6 gas sample extraction ports (one at each axial

thermocouple location). A hydrogen torch is used to ignite gas-phase combustion. This torch remains on during rig stabilization (to ensure safe burnout of all fuel prior to the rig exhaust, especially important if the catalytic reactor has not yet lit off), but is turned off prior to obtaining steady-state data.

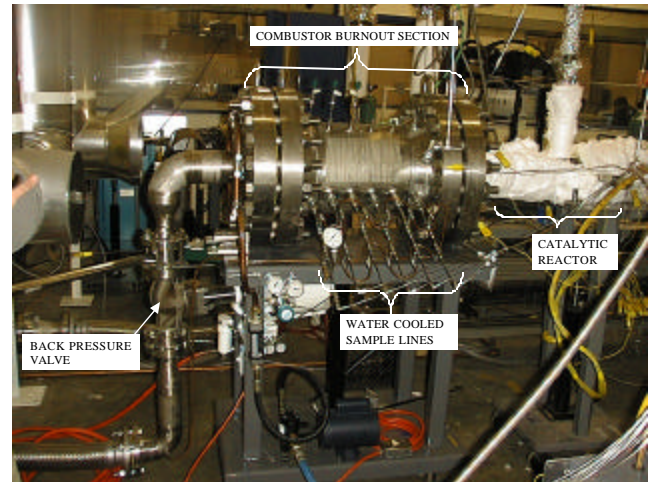


Figure 3. Photograph of PCI's 10 atm sub-scale combustor rig for syngas combustion.

High-pressure air is supplied to the rig from compressors, which can deliver 0.12 pps air at about 145 psia into the rig. At this flow rate, the rig inlet air can be heated to 932 F (500 C). Fuel and diluent are supplied from bottles or Dewar flasks at high pressure, and are pressure regulated to the proper delivery pressure to the rig. All flows (air, fuel, and diluent) are metered with electronic mass flow controllers. Each fuel component is separately metered and then mixed with the other components. For the current tests, five fuel components can be introduced: H_2 , CO , CH_4 , CO_2 , and N_2 .

OPERATING CONDITIONS AND STARTUP PROCEDURES

Tests were conducted at 10 atm, 750 F (400 C) combustor inlet temperature for air and 570 F (300 C) for fuel, and operating conditions were determined from references [3, 4, 5] to simulate F-engine operation on syngas. Note that all emissions reported in ppm are corrected to 15% excess oxygen, dry.

Startup was best accomplished by bringing the reactor to fuel-rich conditions using methane fuel, with some diluent addition to ensure proper mixing. When necessary, a small amount of H_2 was temporarily added to the methane to light off the reactor. Once the catalyst and combustor were lit and the rig was thermally stable, syngas fuel flow was ramped up while methane fuel flow was ramped down, holding catalyst equivalence ratio approximately constant. This startup procedure was economical and safe: it minimized the use of high-volume (costly) laboratory syngas fuel blend, and also avoided use of H_2 during transient and ignition events, where there was a concern that unburned H_2 might enter the exhaust stack and create an explosion hazard. This procedure is similar to syngas combustor startup in actual engine applications.

RCL⁰ SYNGAS TESTING

Results from this study demonstrate the feasibility of using an RCL reactor for syngas combustion and to subsequently obtain ultra-low emissions in the downstream combustor. For the high pressure sub-scale tests, "baseline" operating conditions were based on the IGCC plant at Tampa Electric's Polk Power Station. The Tampa Polk plant operates a GE 107FA combined cycle system on syngas generated from a Texaco oxygen-blown coal gasifier. The current practice of nitrogen injection for NOx control reduces the effective heating value of the fuel.

Fuel Composition

The baseline syngas fuel composition was derived from data gathered in references [3, 4, 5]. The final syngas formula used for baseline testing is tabulated in Table 1. Parametric tests were conducted by keeping the H₂/CO ratio constant while varying the Btu content (diluent content).

H ₂ (%)	CO (%)	CO ₂ (%)	N ₂ (%)	LHV (Btu/ft ³)
20	20	10	50	117

Table 1. Simplified baseline syngas composition used for high-pressure tests.

Preliminary Testing at Atmospheric Pressure

Syngas fuel tests were first performed at atmospheric pressure using a variant of the RCL two-stage (catalytic/gas phase) combustion process originally developed for natural gas. These preliminary tests were intended to provide some initial experience in syngas fuel operation, and in catalyst and combustor behavior using syngas fuels. The results were used to help guide reactor design and test planning for the subsequent high-pressure tests.

Thus, the following atmospheric pressure test objectives were established to facilitate successful high pressure (10 atm) testing:

1. Characterize RCL catalyst lightoff and extinction temperature for syngas fuel.
2. Characterize RCL catalyst operating temperature and reactor fuel conversion.
3. Obtain preliminary NOx and CO emissions.
4. Establish a Standard Operating Procedure (SOP) for rig operation using syngas fuel (e.g. startup, catalyst lightoff, etc.).

Testing and Results. Although the equivalence ratio was varied during atmospheric testing, the syngas blend remained fixed (consistent H₂/CO ratio). Emissions measurements were obtained over a range of conditions, as tabulated in Table 2. Overall equivalence ratio was measured at the emissions probe, downstream of the catalyst and near the exit of the gas-phase combustor. In general, the low emissions measured show that at atmospheric pressure, NOx emissions less than 3 ppm (0.01 lbs/MMBtu NOx) were easily achieved for equivalence ratios as high as 0.53.

From the atmospheric tests, many observations were made and were useful during high pressure testing. For fuel-rich conditions, syngas lightoff temperature was about 356 F (180 C), while extinction temperature was < 176 F (< 80 C). Catalyst

operating temperature and axial profile were similar to those obtained using methane fuel.

Adiabatic Flame Temperature (°C / °F)	Overall equivalence ratio	CO (ppm)	NOx (ppm)
1450 / 2642	0.53	1.5	2.6
1412 / 2573	0.50	0.8	2.4
1300 / 2372	0.45	0.8	1.9
1274 / 2325	0.40	1.4	1.5

Table 2. Summary of emissions measurements at atmospheric pressure.

Testing at High Pressure (10 atm)

Objectives. The primary goal of the sub-scale high-pressure tests was to evaluate emissions performance of the RCL combustion system with syngas fuel. Thus, the primary objectives listed below relate to performance of the downstream combustor during operation on syngas fuel:

1. Characterize combustor emissions (NOx, CO, and UHC) and lean blowout (LBO) at baseline conditions. Vary fuel flow to establish low-emissions turndown range (low NOx and low CO). Use baseline syngas fuel composition, baseline reactor configuration (fixed percentage of air to fuel-rich fuel/air mixture), and baseline (base load) inlet air conditions (constant pressure, air flow, and temperature).
2. Characterize combustor performance (emissions and LBO) for non-baseline syngas fuel compositions. In particular, keep constant H₂/CO ratio but vary Btu content. For each fuel composition tested, vary fuel flow to establish low-emissions turndown range.

Emissions Performance. Emissions measurements reported were obtained from the gas sample port located 15 inches downstream of the catalyst, corresponding to 50 ms residence time. This represents the maximum residence time expected in a low-emissions gas turbine combustor, and therefore also represents the maximum expected NOx emissions for a given operating condition.

All measurements were made with a combustor inlet air temperature of 750 F (400 C) and a syngas fuel temperature of 570 F (300 C). Adiabatic flame temperatures were calculated based on fuel/air ratio as measured by the emissions analyzers (i.e. from gas samples extracted at the 15-inch gas sample probe location).

Figure 4 plots measured NOx and CO emissions as a function of adiabatic flame temperature at 10 atm pressure for a "baseline" syngas composition of 20% H₂, 20% CO, 10% CO₂, and 50% N₂, giving a LHV of 117 Btu/ft³. With this fuel composition, NOx emissions were 2 ppm (0.011 lbs/MMBtu) at the 2550 F (1399 C) flame temperature data point corresponding to the baseline IGCC firing temperature and representing operation at 100% load.

As the fuel/air ratio was decreased, CO emissions remained near zero for flame temperatures greater than about 2250 F (1232 C), permitting a 300 F (149 C) turndown in flame temperature from the 2550 F (1399 C) baseline point, allowing ultra low emissions operation over a wide range of loads. These results –

CO near zero, and NO_x equal to or less than 2 ppm (0.011 lbs/MMBtu) for full load and below – easily met the emissions goal.

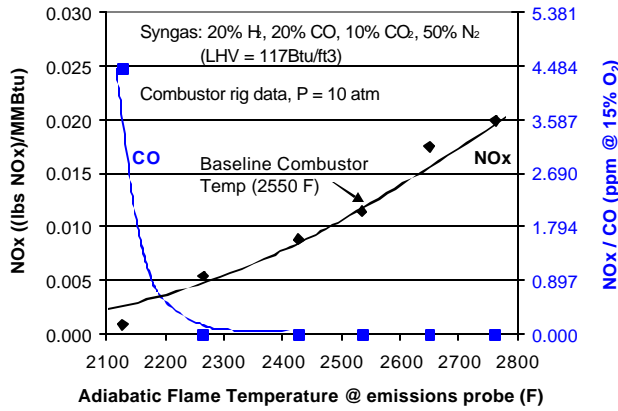


Figure 4. Measured NO_x and CO emissions in PCI's sub-scale rig at 10 atm pressure, as a function of adiabatic flame temperature at the emissions probe.

In another parametric test, heating value of the diluted syngas fuel was reduced to determine operability. NO_x emissions are shown in Figure 5. It is important to note that the right-hand vertical axis in Figure 5 (NO_x values in ppm) is only applicable to the baseline syngas composition, as marked. For the fuel composition with a lower heating value, NO_x emissions in ppm are slightly lower than shown (here, 0.011 lbs/MMBtu is equivalent to 1.6 ppm).

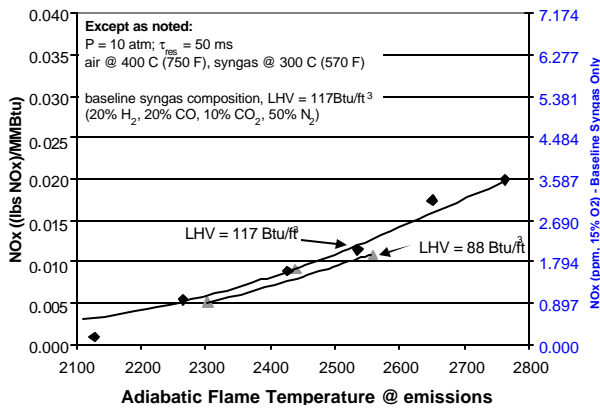


Figure 5. Measured NO_x emissions in PCI's sub-scale rig for two different syngas compositions having LHV's of 88 and 117 Btu/ft³.

It is worth noting that, as shown in Figure 5, catalytic combustion allows stable operation with low emissions for the low Btu syngas case (88 Btu/ft³) even at flame temperatures as low as 2300 F (1260 C). CO emissions were less than 5 ppm in all cases, and were near zero for flame temperatures greater than 2200 F (1204 C). The fuel compositions for the data shown in Figure 5 are listed in Table 3.

H ₂ (%)	CO (%)	CO ₂ (%)	N ₂ (%)	LHV (Btu/ft ³)
15	15	10	60	88
20	20	10	50	117

Table 3. Syngas compositions for data shown in Figure 5, arranged by heating value.

RCL[®] TESTING USING ALTERNATE FUELS

In addition to syngas experiments, tests were performed to observe RCL performance with other low heating value and high-hydrogen fuels. Two additional fuels that were tested were blast furnace gas and refinery fuel gas.

Blast Furnace Gas

RCL combustion of an 82 Btu/ft³ blast furnace gas was tested using the same sub-scale high-pressure combustion rig as that of the syngas. For these tests the simulated blast furnace gas comprised 23% CO, 22% CO₂, 1.4% H₂, 0.6% CH₄, and 53% N₂, and entered the reactor after being heated to about 446 F (230 C). Combustion air (including catalyst-bound or catalyst-side air) entered the reactor at about 662 F (350 C). Results show that combustion of this gas is extremely stable following fuel-rich catalytic reaction, even at adiabatic flame temperatures as low as 2250 F (1232 C).

The high diluent fraction of the fuel means that high fuel-lean equivalence ratios are needed in the combustor burnout zone to achieve the desired flame temperature for the turbine. Tests were performed over a range of adiabatic flame temperatures in the combustor burnout section, from about 2250 to 2500 F (1232 to 1371 C) (representing maximum fuel flow capability of the rig for this blast furnace gas composition). Note that the stoichiometric flame temperature for this blast furnace gas is only about 2700 F (1482 C) for the inlet temperatures tested.

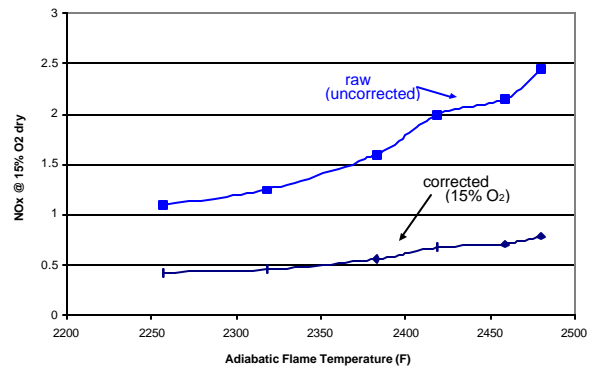


Figure 6. NO_x emissions, uncorrected and corrected to 15% O₂, as a function of adiabatic flame temperature in the downstream combustion zone of the RCL reactor burning blast furnace gas (23% CO, 1.4% H₂, 0.6% CH₄, 22% O₂, and 53% N₂).

For blast furnace gas operation, ultra-low emission levels were achieved for all conditions tested. NO_x emissions for blast furnace gas operation are plotted in Figure 6, as measured by sample extraction from a cooled probe located 15 inches downstream of the catalyst exit. For all conditions tested, NO_x emissions were measured below 2.5 ppm on a raw basis

(uncorrected) and below 1 ppm corrected to 15% O₂ dry. CO emissions were near zero (< 1 ppm) for all conditions shown.

Measured oxygen concentrations following fuel-lean burnout were low as a result of the high level of diluent in the blast furnace gas, varying between about 2.5 and 5.5% for the conditions shown. Thus, the standard correction to 15% O₂ may be misleading since oxygen levels would never reach 15% in an actual engine application. The raw NO_x data are probably as relevant as the corrected NO_x data, or perhaps more so. However, in general, NO_x emissions were ultra-low as a result of the low blast furnace gas flame temperatures.

Refinery Fuel Gas

Testing of RCL combustion of refinery fuel gas was also conducted using the same hardware configuration as previous tests for syngas and blast furnace gas. Results showed NO_x emissions below 3 ppm for flame temperatures below 2800 F (1538 C).

For the refinery fuel gas tests, the simulated refinery fuel gas comprised 30% H₂ and 70% CH₄, and entered the reactor without passing through a fuel heater. However, some fuel heat was obtained from hot combustor rig components so that the fuel plenum gas temperature measured about 347 F (175 C). Combustion air entered the reactor at about 734 F (390 C).

Tests were performed over a range of adiabatic flame temperatures, from about 2400 to 3000 F (1316 to 1649 C) in the combustor burnout section, and at a pressure of about 10 atm. NO_x and CO emissions were measured at each condition, as well as O₂ and CO₂.

NO_x emissions for the RCL combustion of refinery fuel gas are plotted in Figure 7, as measured by the downstream emission probe. NO_x emissions were measured below 3 ppm for flame temperatures less than about 2800 F (1538 C). CO emissions were less than about 1 ppm for all conditions shown.

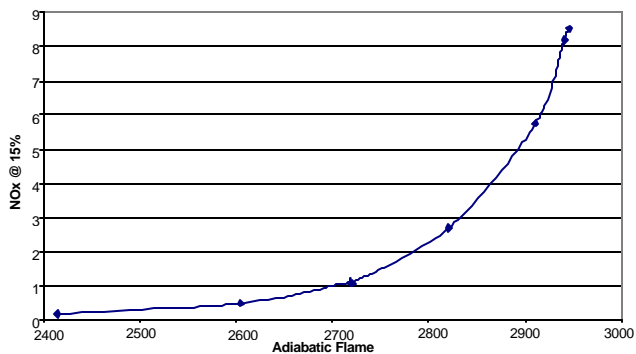


Figure 7. NO_x emissions, corrected to 15% O₂, as a function of adiabatic flame temperature in the downstream combustion zone burning refinery fuel gas (30% H₂ and 70% CH₄).

CONCLUSIONS

Successful operation of the RCL combustion system was demonstrated at 10 atm for syngas fuel. NO_x emissions were measured in the range of 0.01 lbs/MMBtu (< 3 ppm) and were below this value under modified operating conditions during parametric testing simulating operation of Tampa Electric's IGCC plant.

In addition to the syngas fuel tests, further tests were conducted with alternate fuels with low heating values and/or

high hydrogen content. The two types of alternate fuels tested were blast furnace gas and refinery fuel gas. Both fuels performed well, with good flame stability for blast furnace gas with NO_x levels below 2.5 ppm and refinery fuel gas with NO_x levels below 3 ppm.

ACKNOWLEDGMENTS

Precision Combustion, Inc. would like to thank the U.S. Department of Energy-Fossil Energy program for their support and funding throughout the duration of this project. In particular, PCI would like to extend their gratitude to Mr. Chuck Alsup, Mr. Rich Dennis, Mr. Jose Figuera, and Dr. Geo Richards for their encouragement and support in the development of this technology.

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