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CATALYTIC COMBUSTION ENABLING TECHNOLOGIES DEVELOPMENT PROGRAM FOR INDUSTRIAL GAS TURBINE SYSTEMS

Final Report

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DISCLAIMER

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1. SUMMARY

PCI has successfully completed this enabling technologies development program, leading to development of a robust, low-lightoff-temperature, ultra-low-emissions gas turbine catalytic combustion system burning natural gas. Collaboration with a major OEM industrial gas turbine manufacturer, Solar Turbines Inc., provided technical program direction and high-pressure (15-17 atm) test support.

Sub-scale durability testing at 9 atm demonstrated 1000 hours of operation without degradation in catalyst performance. In addition, full-scale catalyst modules were fabricated and successfully tested at full pressure (15-17 atm) at Solar Turbines, achieving NOx < 3 ppm with CO < 10 ppm at simulated Solar Taurus 70 engine conditions, exceeding ATS emissions goals by a wide margin. At these ultra-low emissions levels, combustion-driven pressure oscillations (CDPO) were less than 0.15% peak-to-peak (0.35 psi peak-to-peak).

2. MAJOR PROGRAM ACCOMPLISHMENTS

A new approach to catalytic combustion was developed under the present program, offering significant advantages as compared with conventional lean-premixed catalytic combustion. PCI has termed this new approach Rich-Catalytic / Lean-burn (RCL) combustion. Its major advantages are operation without a preburner (low lightoff and extinction temperatures), avoidance of issues of auto-ignition and flashback, and long catalyst life (non-oxidizing fuel-rich catalyst environment) -- all achieved with ultra-low NOx (< 3 ppm) performance.

The specific major accomplishments under this program were:

1. Development of PCI's Rich-Catalytic / Lean-burn (RCL) approach to catalytic combustion, with its significant operational advantages as compared to previous systems;

2. Full-scale RCL catalytic combustion tests at Solar Turbines' high-pressure facility, demonstrating robust catalyst operation and ultra-low emissions (NOx < 3 ppm, CO < 10 ppm) at simulated Solar Taurus 70 engine conditions;

3. Catalyst formulation development and testing at sub-scale; and 1000 hours of high-pressure sub-scale catalyst durability testing, without performance degradation;

4. Several patent applications were submitted relating to the RCL concept and its application.

3. STATEMENT OF WORK

The purpose of the present program was to develop catalytic combustion technologies designed for insertion into an Advanced industrial gas turbine engine, with technical potential for achieving NOx < 5 ppm with acceptable CO and UHC, 8000 hours durability, and < 15% cost add on. Based on discussions with Solar Turbines and DOE, the Solar Taurus 70 engine was selected as an appropriate and commercially significant Advanced industrial gas turbine engine. Based on testing at Taurus 70 engine conditions, the technical objectives of this program were met, as will be described in detail in this report.

The milestones for 1000-hour catalyst durability testing and high pressure full scale module combustor tests at an OEM industrial gas turbine engine manufacturer's site were successfully met. These milestones were completed under the present program, as will be described in detail in this report.

4. BACKGROUND – MOTIVATION AND RCL DEVELOPMENT

4.1. ROLE OF CATALYSIS IN LOW-EMISSIONS COMBUSTION

By catalytically pre-reacting a portion of the fuel/air mixture destined for a combustion zone, lean stable combustion can be obtained with significantly leaner mixtures than is otherwise possible. As a result, low CO and UHC emissions can be obtained with low peak flame temperatures (as low as 1300 C (2370 F)), with low single digit NOx emissions.

The earliest work on what is now termed catalytic combustion was conducted by PCI's Chief Scientist Dr. William Pfefferle (1974) while at Engelhard. The original catalytic combustor (Pfefferle, 1974) is a ceramic honeycomb monolith of nested catalytically-coated parallel channels placed within a combustion chamber. In this original-type catalytic combustor, surface reactions release heat and radicals into the boundary layer above the surface, and the surface operates under mass transfer limitation at the adiabatic flame temperature. Catalytic combustion increases the mixture reactivity by achieving partial reaction at the catalytic surface, adding both heat and reactive intermediates to the gas phase. As a consequence, reactor operation can be at lean limits well beyond those feasible without the influence of the catalyst. Early work on systems of this type were conducted at Engelhard (Pfefferle, 1974), Acurex (Kesselring, 1979), Westinghouse (Pillsbury, 1984), NASA (Anderson, 1975), the Air Force (Rosfjord, 1976), and elsewhere.

Active interest in catalytic combustion revived during the early 1990s, as it became clear that continued pressure for reduced emissions could not likely be met entirely by re-design of conventional combustors. A new approach of partial conversion in the catalyst bed, and the use of metal substrates with innovative catalyst and system design to circumvent material issues of shock resistance and non-availability of reliable high temperature catalysts revived catalytic combustion for power generation. Metal-substrate type catalyst beds have been employed for catalytic combustion with increasing success demonstrating the low NOx potential of catalytic combustion for ground power generation (Dalla-Betta, 1997; Dutta, 1997; Smith, 1997; Pfefferle, 1996, 1997).

4.2. CHALLENGES TO CATALYTIC COMBUSTION

In early gas turbine catalytic combustion systems (Pfefferle, 1974) catalytic and gas-phase combustion were combined into a single stage, with the catalytic reaction stabilizing (inducing) immediate gas-phase reactions. Thus, complete combustion of the fuel/air mixture occurred within the catalyst bed. A major limitation of this single-stage system was the need for catalyst and substrate materials capable of withstanding the maximum combustion temperature. Generally, this required ceramic-type substrate materials which were prone to failure by thermal shock, particularly in gas turbine engine applications. In addition, final combustion temperatures were limited by the material temperature limits of substrate and catalyst.

For modern gas turbines, with high turbine inlet temperatures for high efficiency, complete combustion of the fuel/air mixture within the catalyst bed is not generally feasible, due to the mismatch between the high turbine inlet temperatures and the maximum material temperature limits of available catalyst and substrate materials. Thus, so-called hybrid catalytic combustor systems are employed. The hybrid system combines a downstream gas-phase combustion section with an upstream catalytic reactor, both operating at the same fuel-lean equivalence ratio.

By only partially reacting the fuel in the catalyst bed, at moderate temperatures, hybrid systems allow the use of metallic catalyst supports, which have the best mechanical properties for gas turbine engine applications, offering excellent resistance to both thermal and mechanical shock. For long-term durability of modern high-temperature alloys maximum material temperatures are generally less than 900 C (1650 F), while engine firing temperatures typically exceed 1300 C (2370 F) at full load. This mismatch in temperature requirements dictates that the zone of combustion completion (burnout zone) must be physically separated from the catalyst bed, with catalyst temperatures maintained below material limits. Therefore, only a fraction of the fuel's heat of reaction can be liberated in the catalyst bed.

Thus, the success of the hybrid system is dependent upon the catalytic reactor's ability to limit reactions (and thus temperature), so that the metal substrate within the reactor may operate below its maximum material temperature limit. The degree of reaction can be limited by chemical reaction rate upon the catalyst, by mass transfer of reactants to the catalyst, or by channeling within the reactor such that only a limited fraction of the fuel can contact the catalyst. In all cases, however, it is imperative that uncontrolled gas-phase reactions do not occur within the catalytic reactor, since this implies a loss of reaction limitation and ultimate over-temperature and failure of the catalyst bed.

4.3. RCL Development and Advantages

Under the present program, a new catalytic reactor concept was developed to provide definitive limitation of reactions, resolving and precluding issues of over-temperature and failure of the catalyst bed from flashback or auto-ignition. This new concept, which we call Rich-Catalytic / Lean-burn (RCL) combustion, contacts a fuel-rich mixture with the catalyst, and uses the remaining combustion air (not yet mixed with fuel) to provide catalyst cooling.

A schematic of the RCL system is shown in Figure 4.3.1. As shown, the combustion air stream is split into two parts upstream of the catalyst: one portion is mixed with all of the fuel and contacted with a catalyst, while a second portion is used to backside cool the catalyst. At the exit of the reactor, the catalyzed fuel/air stream and the cooling air are rapidly mixed to produce a fuel-lean, reactive mixture prior to final combustion.

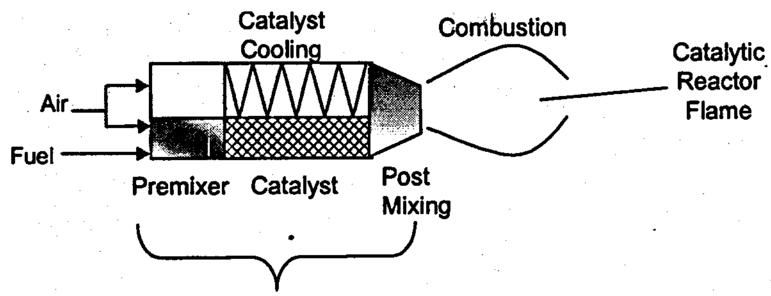

Figure 4.3.1. *Schematic of RCL system. A fuel-rich fuel/air mixture contacts the catalyst, while heat is extracted into a cooling air stream. The cooling air stream and the catalyzed stream are rapidly mixed downstream of the catalyst, but prior to final combustion, to create a fuel-lean fuel/air mixture for the low-NOx burnout zone.*

By passing all of the fuel over the catalyst, the catalyst cooling stream remains free of fuel, precluding failure by flashback or auto-ignition to the cooling stream. At the same time, the fuel-rich mixture contacting the catalyst has insufficient oxygen to completely oxidize all of the fuel, thus limiting the extent of catalyst-stage reaction and enabling limitation of the catalyst-stage operating temperature to a safe value.

The RCL system thus provides significant operational advantages. Most notably, the RCL reactor requires no preburner, is immune to issues of auto-ignition and flashback, and provides long catalyst life (as a result of the non-oxidizing fuel-rich catalyst environment), while providing ultra-low NOx (< 3 ppm) performance.

In summary, RCL provides the following advantages:

- No preburner – space, cost and durability advantage.
- Integrated compact premixer using simple existing technology.
- Compact - capable of fitting to existing engine envelopes.
- High firing temperature operation ideal for ATS applications.
- Robust operation, avoiding catalyst failure by flashback/autoignition.
- Long life due to fuel-rich catalyst environment and moderate wall temperatures.
- Simple control system.

The RCL concept has been patented with rights granted to the DOE.

5. WORK PERFORMED AND RESULTS ACHIEVED

Because of the significant advantages offered by Rich-Catalytic / Lean-burn (RCL) combustion, efforts under the present program were focused early in the program toward continued RCL development and definition for industrial gas turbine engine applications, specifically for Solar Turbines' conditions. This report describes the work performed and results achieved regarding the basic enabling technologies underlying the RCL system, as well as complete RCL combustor integration and demonstration at industrial gas turbine engine conditions.

5.1. Catalyst Development and Testing (Sub-Scale, High-Pressure)

Catalyst formulations which passed preliminary screening in a simple coupon test rig were applied to a sub-scale high-pressure RCL reactor for evaluation. Performance indicators of particular interest at high pressure were catalyst lightoff and extinction temperature, degree of conversion of fuel and oxygen within the reactor, catalyst operating temperatures, and product composition.

Tests were performed in a high-pressure sub-scale rig at pressures from 9 to 15 atm. For natural gas fuel having one or two percent ethane, PCI's catalysts typically light off in the vicinity of 300 C. For natural gas fuel with greater than two percent ethane (or higher-order hydrocarbons) lightoff can occur at inlet temperatures below 280 C. This is shown in Figure 5.1.1 below, which indicates a lightoff temperature between about 260 and 280 C on natural gas fuel, at 15 atm pressure. In Figure 5.1.1, inlet gas temperature, catalyst surface temperature, and gas temperature exiting the module (following mixing of the catalytically reacted stream with the catalyst cooling air stream, but prior to gas-phase combustion) are plotted as a function of time in minutes. Lightoff occurs when the heat of reaction results in an increase in catalyst operating temperature and catalyst exit temperature as compared to the gas inlet temperature.

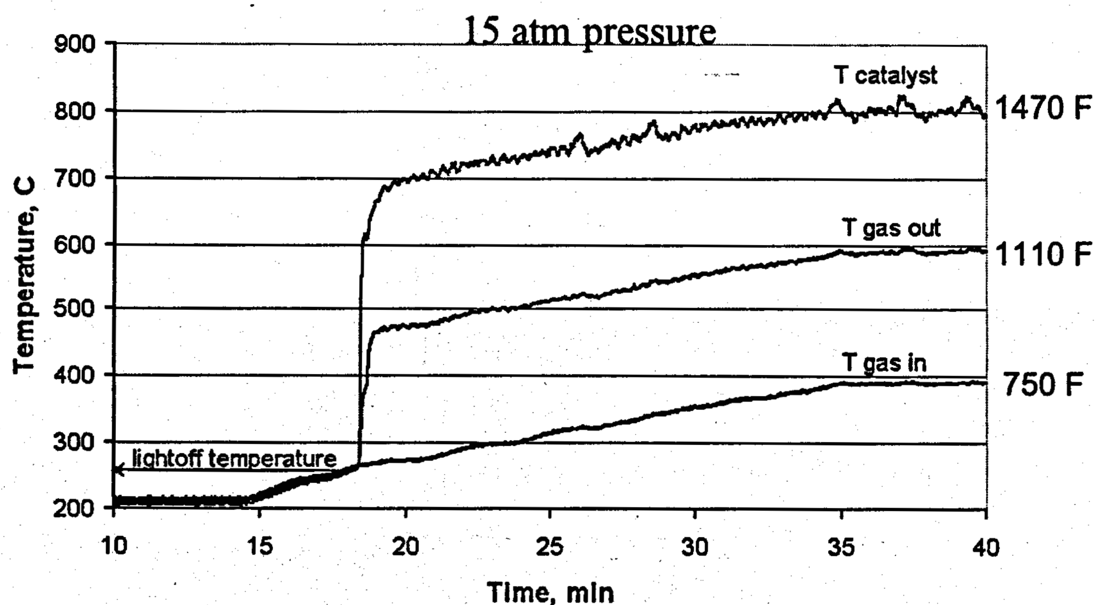

Figure 5.1.1. Catalyst lightoff in a sub-scale high-pressure (15 atm) RCL reactor operating on natural gas fuel. Inlet gas temperature ("T gas in"), catalyst surface temperature ("T catalyst"), and gas temperature exiting the module ("T gas out") are plotted as a function of time in minutes.

Following catalyst lightoff, the inlet air temperature can be reduced well below the initial lightoff temperature without extinguishing the catalyst. Thus, once lit (active), the catalyst remains lit (active) down to inlet temperatures approaching ambient. Following the catalyst lightoff event depicted in Figure 5.1.1, the inlet air temperature was reduced to less than 200 C, but catalyst activity was not diminished. This is shown below in Figure 5.1.2, which plots the same parameters as Figure 5.1.1, now after several hours of durability testing following the initial lightoff. Here, still at 15 atm pressure and with the same flow of natural gas fuel, catalyst activity was maintained until the fuel was shut off at an inlet air temperature less than 200 C.

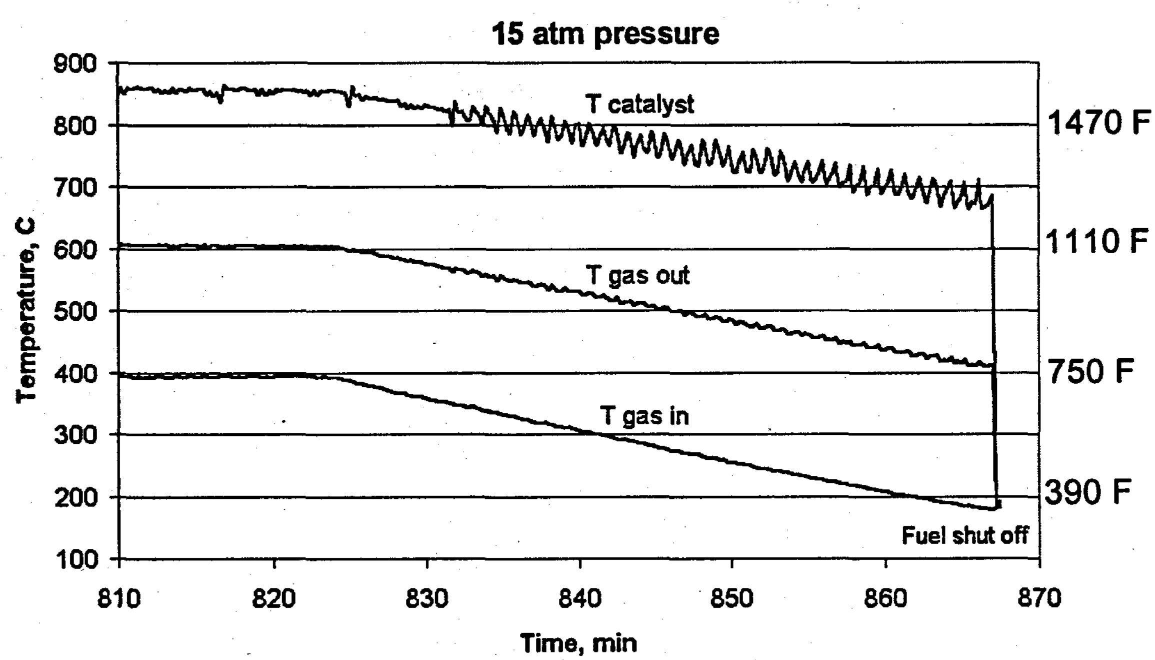

Figure 5.1.2. Catalyst extinction does not occur until the fuel is shut off at an inlet air temperature less than 200 C. Data were obtained for the same sub-scale high-pressure (15 atm) RCL reactor for which data were shown in Figure 5.1.1. Again, inlet gas temperature ("T gas in"), catalyst surface temperature ("T catalyst"), and gas temperature exiting the module ("T gas out") are plotted as a function of time in minutes.

Under the present program, three activities were conducted for development of catalyst and substrate formulations for PCI's RCL reactor. These were:

1. Trials of new formulations and processes in the chemistry laboratory (e.g. adhesion, materials and process compatibility, catalyst and substrate microstructural characterization),
2. Catalyst activity screening on thin strips (or "coupons"), and
3. Extensive catalyst activity and performance testing in a sub-scale RCL reactor at both atmospheric pressure and 10-15 atm pressure conditions.

For catalyst screening, catalyst/substrate combinations are applied to flat, thin strips (or "coupons") for rapid testing. The coupons are installed in a simple test rig, and exposed to a flow of premixed, preheated fuel and air. Catalyst performance is evaluated by the surface temperature rise due to catalytic surface reactions, as measured by a type K thermocouple contacting the coupon. Temperature rise is recorded as a function of inlet fuel/air temperature and mixture ratio. Both as-prepared and furnace-aged coupons are measured and the results are compared with bench combustor performance. Gas chromatograph (GC) analysis of the pre- and post-catalyst streams is also obtained, to provide a measure of fuel conversion and reaction rate. Following coupon testing, promising catalysts are tested at RCL operating conditions, in a sub-scale reactor at both atmospheric pressure and high (10-15 atm) pressure.

Catalyst screening results for five leading catalyst formulations are shown in Figure 3.2.1. Here, the activity level of five catalyst formulations (labeled "A" through "E") are compared on the basis of catalyst temperature (black, labeled "Tsurf"), lightoff temperature (blue, labeled "T l-o"), and fuel conversion (green, labeled "conv").

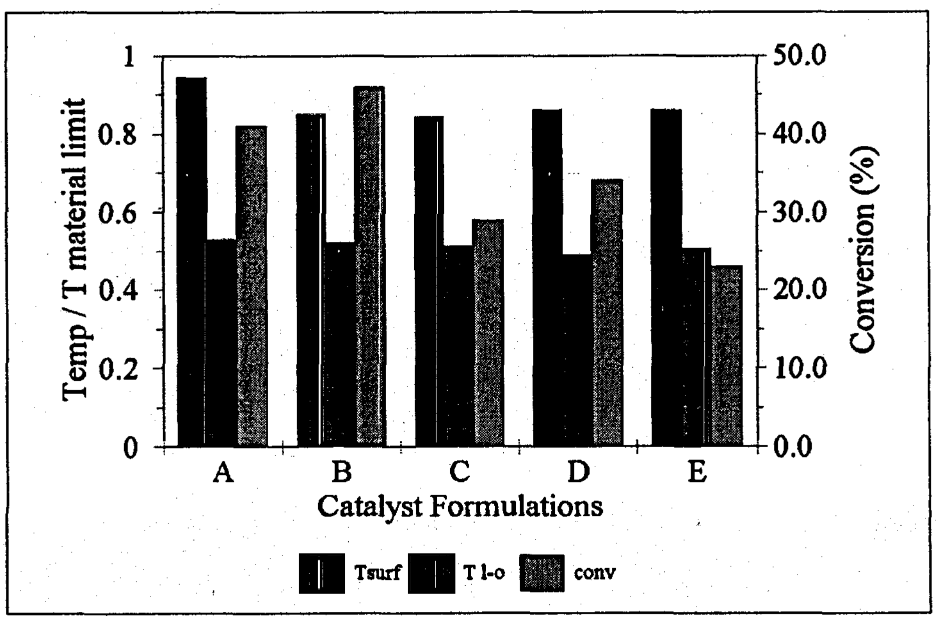

Figure 3.2.1. Comparison of catalyst screening data obtained for five catalyst formulations (labeled A through E) at RCL operating conditions.

Extensive testing was performed at high pressure (10 atm) on three catalyst formulations, in a sub-scale RCL reactor. Temperature profiles axially through the reactor (stations 1 through 9) are shown for these three catalyst formulations in Figure 3.2.2. In general, surface temperatures were moderate and within the material limit temperature, with catalyst formulation "C" showing the best performance in terms of steady well-moderated operating temperatures throughout the full length of the RCL reactor.

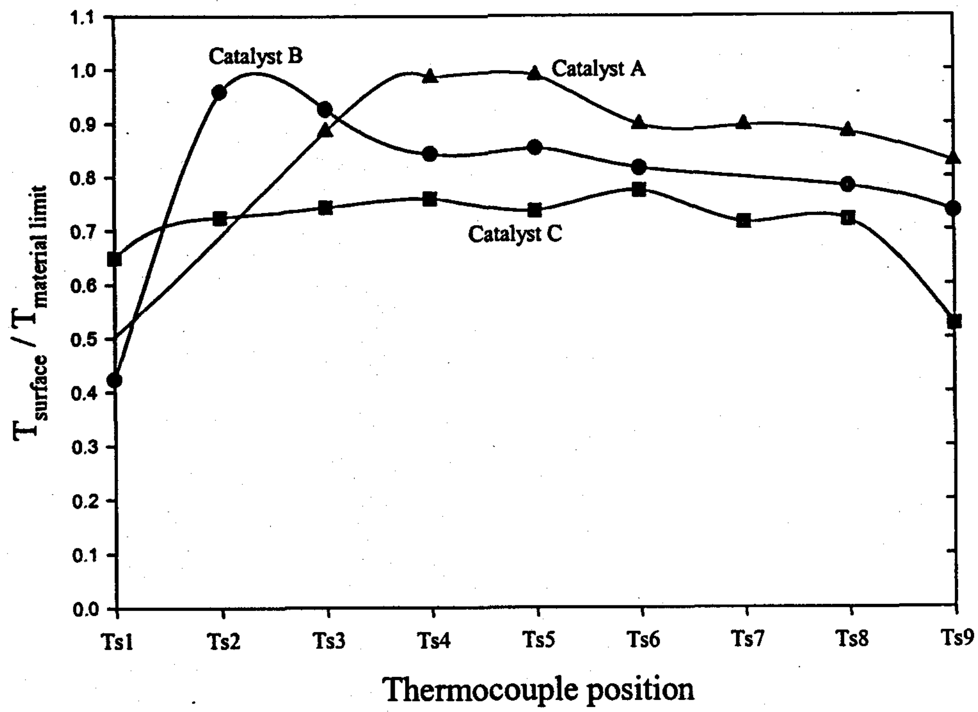

Figure 3.2.2. Catalyst temperature profiles in RCL reactor operating at 10 atm pressure, for A, B, and C catalyst formulations.

5.2. 1000-HOUR CATALYST DURABILITY TESTING

A high-pressure sub-scale catalyst durability test rig was fabricated at PCI, and a sub-scale RCL catalytic reactor was successfully durability tested for 1000 hours, at 9 atm pressure, without measurable performance degradation. Gas samples were analyzed in a gas chromatograph periodically throughout the test period. Gas sample analysis, and the gas temperature exiting the module, confirmed that the catalytic reactor showed no measurable loss in fuel conversion and gas temperature exiting the module during the 1000 hours. In this test the inlet temperature was maintained at 500 C to compensate for high heat loss from the sub-scale rig.

Post-test materials analysis indicates negligible loss of catalyst and support during the 1000 hour test. Based on previous 100- and 500-hour tests, it appears that catalyst sintering occurs during the first 100 or 200 hours, and then becomes negligible. With these results, maintained catalyst performance at 1000 hours indicates good technical potential for maintained catalyst performance at 8000 hours as well.

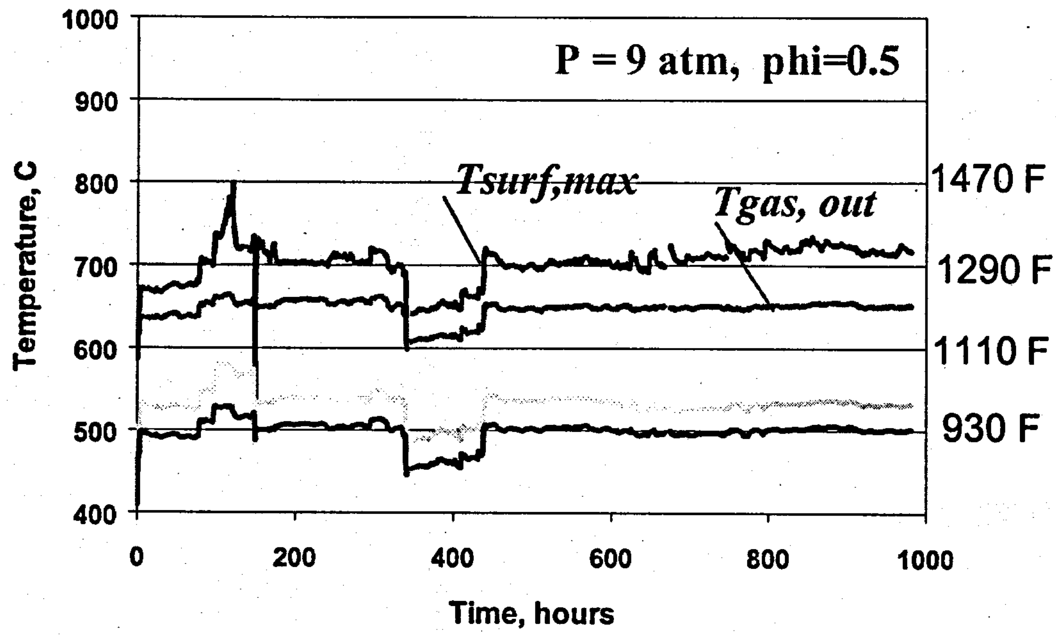

Figure 5.4.1. 1000-hour catalyst durability test, showing no measurable performance degradation. Gas inlet temperature (bottom curve, black), catalyst operating temperature (highest curve, mauve, labeled "Tsurf, max"), and gas temperature exiting the module (magenta curve, labeled "Tgas, out") are plotted against time for 1000 hours.

5.3. Full-Scale, High-Pressure RCL Combustion Tests (At Solar Turbines)

A full-scale RCL module was fabricated for testing in Solar Turbines' high-pressure single-injector combustor test facility. The catalyst bed was sized to replace a single injector (1 of 12) in a Solar Taurus 70 engine.

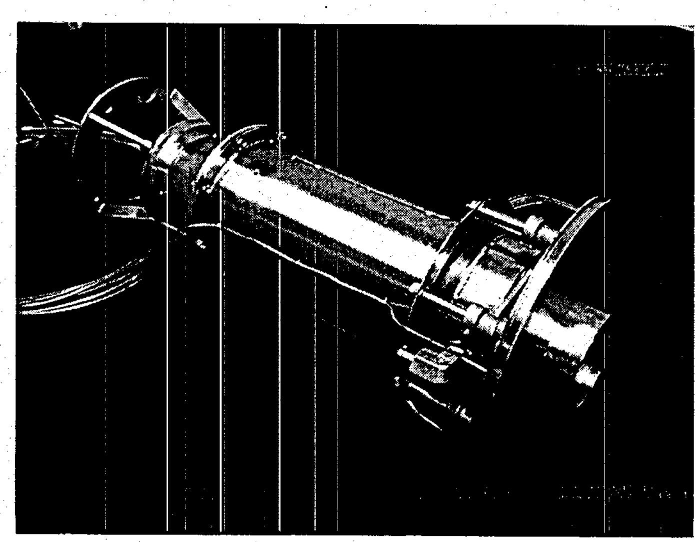

Figure 5.5.1. Photograph of full-scale catalytic combustor module.

A photograph of the module is shown in Figure 5.5.1. Catalytically reacted gases exit the module via a duct visible at the right-hand side of the photograph. For Solar rig testing, this exit duct is fitted into a grommet seal at the upstream end of Solar's rig combustor liner. Gas-phase combustion (burnout) takes place in the combustor liner.

A schematic of the complete assembly, as joined with Solar's rig combustor liner, is shown in Figure 5.5.2. In general, the catalyst is intended to improve combustion stability and turndown at the flame anchor point, but is not necessarily intended to provide gas-phase ignition. Solar's torch igniter was used when needed to ignite gas-phase combustion during rig testing.

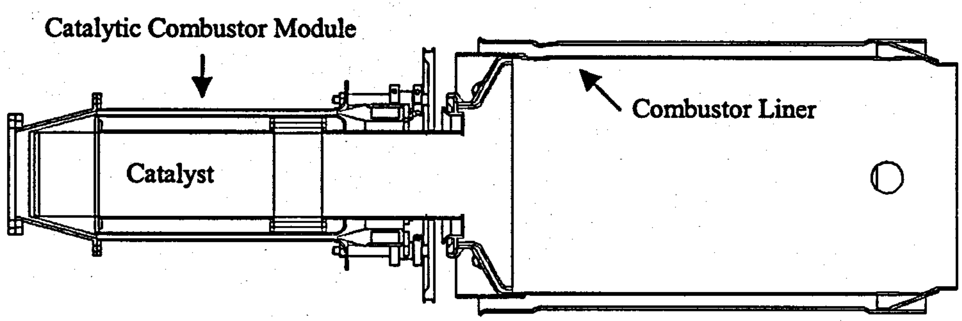

Figure 5.5.2. Schematic of catalytic combustor module coupled with Solar rig combustor liner.

5.3.1. Atmospheric-Pressure Full-Scale RCL Test Results

Initial testing of the full-scale RCL module was performed at PCI, in an atmospheric pressure test rig capable of flowing up to 0.5 pps air at temperatures up to 600 C. The test rig includes a combustor liner downstream of the catalyst, which opens to an atmospheric-pressure exhaust duct. Because the test rig is open to atmosphere, traversable gas sample probes can be inserted from the downstream end to perform mixing surveys of the test hardware (e.g. premixer, catalyst module).

Premixer Verification

To minimize the overall length of the RCL system, the fuel/air premixer for the full-scale module was designed as a reverse-flow annular mixer located on the outside diameter of the catalyst assembly. Prior to module assembly, the premixer was tested alone, without the catalyst, at atmospheric pressure. A gas sampling probe was traversed azimuthally (rotated) in the mixing duct, and methane concentrations were analyzed by gas chromatograph.

Data were obtained at the premixer exit (catalyst inlet) at 40 points around the 360-degree premixer annulus. Methane concentration, normalized by the mean, is shown in Figure 5.5.3 for each of the 40 probe positions. The root-mean-square deviation from the mean (unmixedness) is less than 4%, meeting our design target of <5% (for ultra-low NOx emissions performance). It should be noted that the reactor can handle a higher level of unmixedness, but in order to achieve low single-digit NOx at high firing temperatures, <5% unmixedness was targeted. In Figure 5.5.3 two small peaks are visible, at 120 and 300 degrees, and largely account for the non-zero rms unmixedness. The two peak-fuel locations are opposite each other, and are in-line with the two fuel inlets to the fuel manifold which feeds the premixer. Future hardware adjustments can further reduce the level of these peak values.

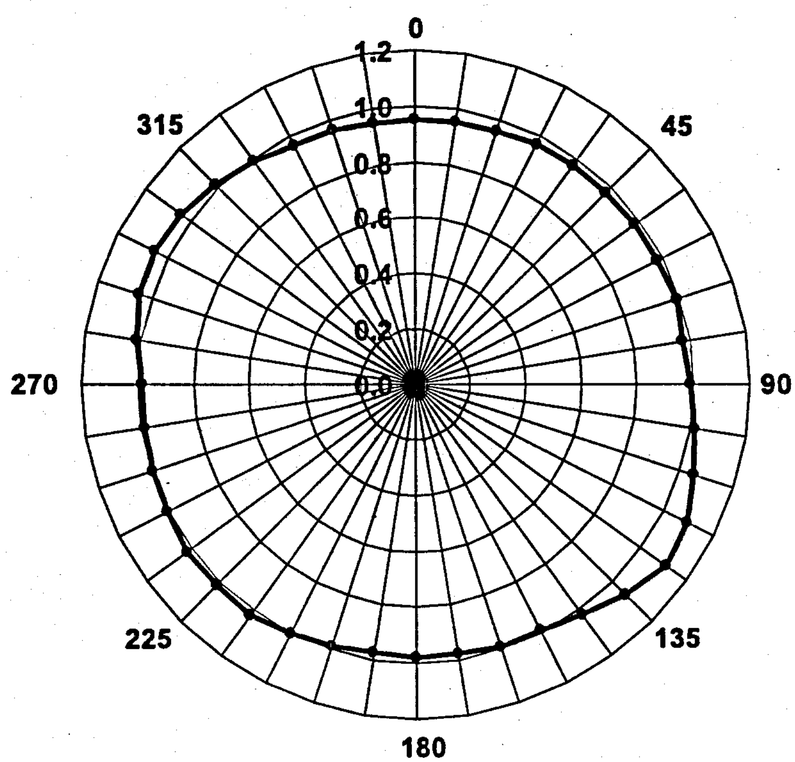

Figure 5.5.3. Radial plot of measured methane concentration, normalized by average, at premixer exit (upstream of catalyst bed).

Post-Catalyst Mixing

After final assembly of the module, methane concentration was measured in the post-catalyst mixing duct, where partially reacted catalyst effluent mixes with catalyst cooling air prior to final fuel-lean burnout. Measurements were made in a cross-stream plane located 4 inches downstream of the catalyst exit, at points spaced 1/2-inch apart in a square grid array, via gas sampling probe and gas chromatograph analysis. Post-catalyst mixing measurements were obtained at atmospheric pressure.

A plot of all measured post-catalyst duct methane concentrations is shown in Figure 5.5.4, normalized by the average concentration in the core flow. The data are plotted against radial distance from the duct centerline. The core flow is considered to include only those data points having a normalized value greater than 0.8 (as seen in Figure 5.5.4, these "core" points are within a 1.2-inch radius of the duct centerline). Near the duct wall, the fuel concentration is weak because excess air was introduced along the wall to ensure that flame propagation from downstream to upstream could not occur in the wall boundary layer. Within the core flow, the data show a 6.4% rms unmixedness (root-mean-square deviation from the core mean), with no significant fuel concentration peaks. This is acceptably close to our 5% rms unmixedness target for low NOx emissions.

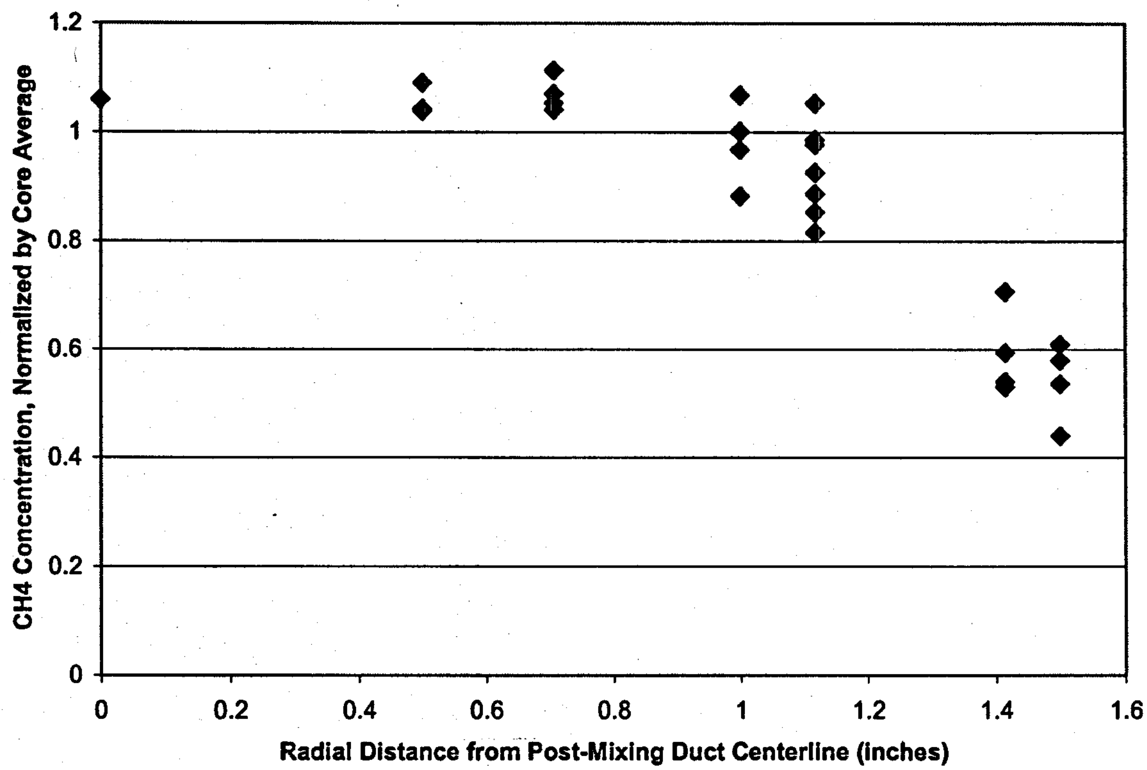

Figure 5.5.4. Methane concentration measurements in post-catalyst mixing duct, normalized by average of core flow (core flow points are less than 1.2 inches from duct centerline).

Pressure Loss and Effective Area of Catalyst Module

De-burred, wall-flush static pressure ports were installed at four (4) locations in the RCL module: 1. at the premix duct exit (just prior to the catalyst); 2. within the catalyst bed, near its upstream end; 3. within the catalyst bed, near its downstream end; and 4. in the post-catalyst mixing duct, just prior to its exit into the downstream combustor liner.

Under cold conditions (no catalyst activity), methane fuel flow was varied from about 15% to 100% of expected operating range (overall equivalence ratio from 0.1 to 0.6 at the exit of the post-catalyst mixing duct), and static pressure measurements were obtained throughout the module. For these tests the combustor liner was open to atmosphere.

Static pressure data are plotted in Figure 5.5.5, in terms of percent deviation from ambient. The shell pressure indicates the static pressure entering the module ("shell" refers to the shell of the test rig). Overall pressure loss is about 3.25% at the conditions tested, irrespective of fuel flow. Losses occur at the catalyst inlet and in the mixing region downstream of the catalyst.

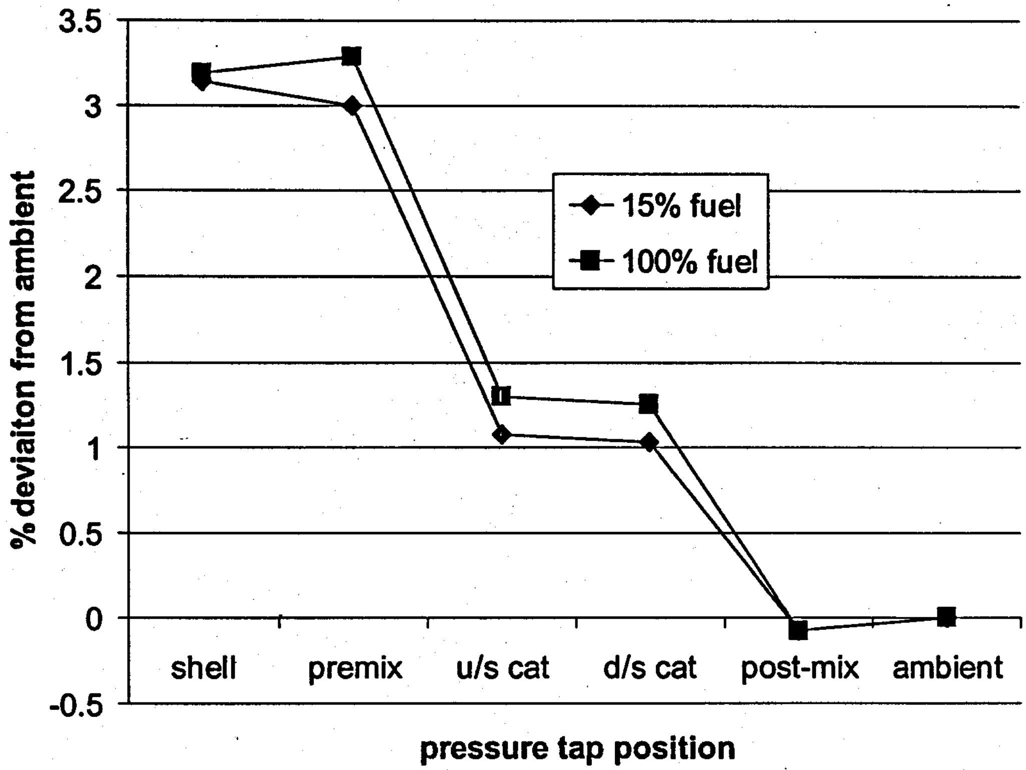

Figure 5.5.5. Static pressure, in percent deviation from ambient, at six stations throughout catalyst bed and rig: shell (plenum), premix duct, upstream end of catalyst, downstream end of catalyst, post-mix duct, and combustor liner (ambient).

At ambient temperature, and with no fuel flowing, the air flow to the RCL module was varied to check module effective area as a function of air flow. The results are shown below, in Figure 5.5.6. For simple orifice losses there should be no dependence of effective area on air flow. The catalyst bed, however, introduces frictional losses, which result in increased effective area (lower percent pressure drop) with increased air flow (and also with increased pressure, although this is not shown here). The solid line in Figure 5.5.6 represents a least-squares-fit to the data. At an atmospheric pressure base-load air flow rate of approximately 0.2 pps, the effective area of the module is 2.1 square inches for the no-fuel, ambient temperature case.

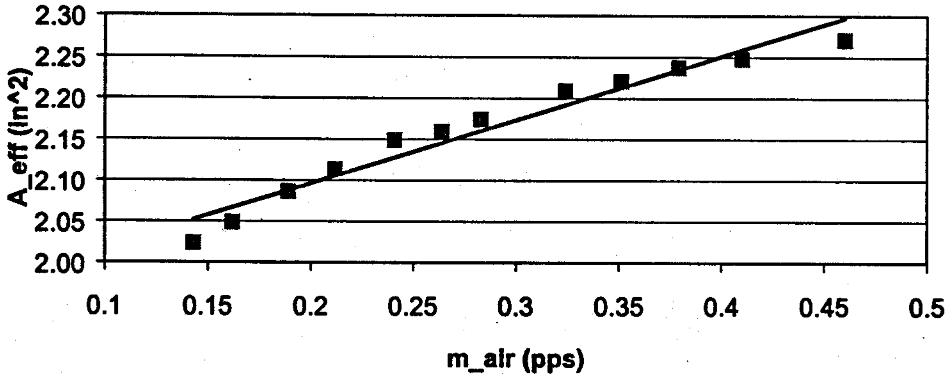

Figure 5.5.6. Effective area of the RCL module, at ambient temperature and with no fuel flow.

5.3.2. High-Pressure Full-Scale RCL Test Results

High-pressure testing of the full-scale RCL module was performed at Solar Turbines. For these tests, a Solar-provided backside-cooled combustor liner was used, of nominally 8-inches in diameter. A water-cooled back-pressure valve downstream of the combustor exit allowed testing at pressures up to 250 psig. An emissions rake at the combustor exit (nominally 25-30 ms combustor residence time) feeds gas samples to an emissions train consisting of analyzers for NOx, CO, UHC, O_2, and CO_2. The UHC sample is not dried, but all other analyzers receive a chiller-dried sample. The NOx analyzer range is 25 ppm at its most sensitive setting, with an accuracy better than 2% of full scale (0.5 ppm). The analyzers are zeroed and calibrated twice each day, and a linearity check is performed on the NOx analyzer monthly, using a range of gases including a bottom-end calibration gas of 5 ppm NO.

Solar's rig combustor liner was modified slightly to provide optimal conditions for post-catalyst burnout with low emissions. Scoops were placed over the dilution air holes to direct dilution air downstream, away from the primary zone. Dome cooling air holes were closed with Ni-Cr tape, to reduce primary-zone dilution by cooling air; this was possible because the non-swirling flow exiting the catalyst does not impinge on the dome or liner wall, greatly reducing heat load. Finally, the combustor liner was given a thermal barrier coating (TBC), for hotter walls and reduced CO wall quenching. The combined catalyst and combustor liner were shown schematically in Figure 5.5.2.

Instrumentation

Thermocouples were welded to the catalyst substrate at nine (9) locations throughout the catalyst bed, to provide a measure of catalyst operating temperature. Thermocouples were also welded to the reactor housing and post-catalyst duct, and to Solar's rig combustor liner. Gas-temperature thermocouples were installed in the premixer and post-catalyst duct.

Gas sampling ports were provided in the following locations, for gas chromatograph (GC) analysis of gas composition: premixer, catalyst bed, post-catalyst duct, and post-combustion duct (emission rake). GC analysis was used to determine air flow splits (and effective areas) at pressure and with reaction, degree of catalytic reaction, and catalyst effluent composition.

Catalyst Performance

With natural gas fuel flowing through the catalyst bed (giving 0.55 equivalence ratio at the post-catalyst mixing duct exit), and with the downstream combustor ignited and providing burnout, the rig inlet air temperature was ramped up from approximately 600 to 800 F (320 – 420 C) at 15 atm pressure. At a temperature just over 600 F (320 C) the catalyst became active, and the catalyst surface temperature and gas exit temperature increased to values well above the 320 C inlet temperature. This event is shown below in Figure 5.5.7, where the values for the maximum catalyst surface temperature and catalyst module gas exit temperature are plotted versus rig inlet air temperature. The data points shown were obtained from the transient data file operating during lightoff, at a data collection rate of one data point per second.

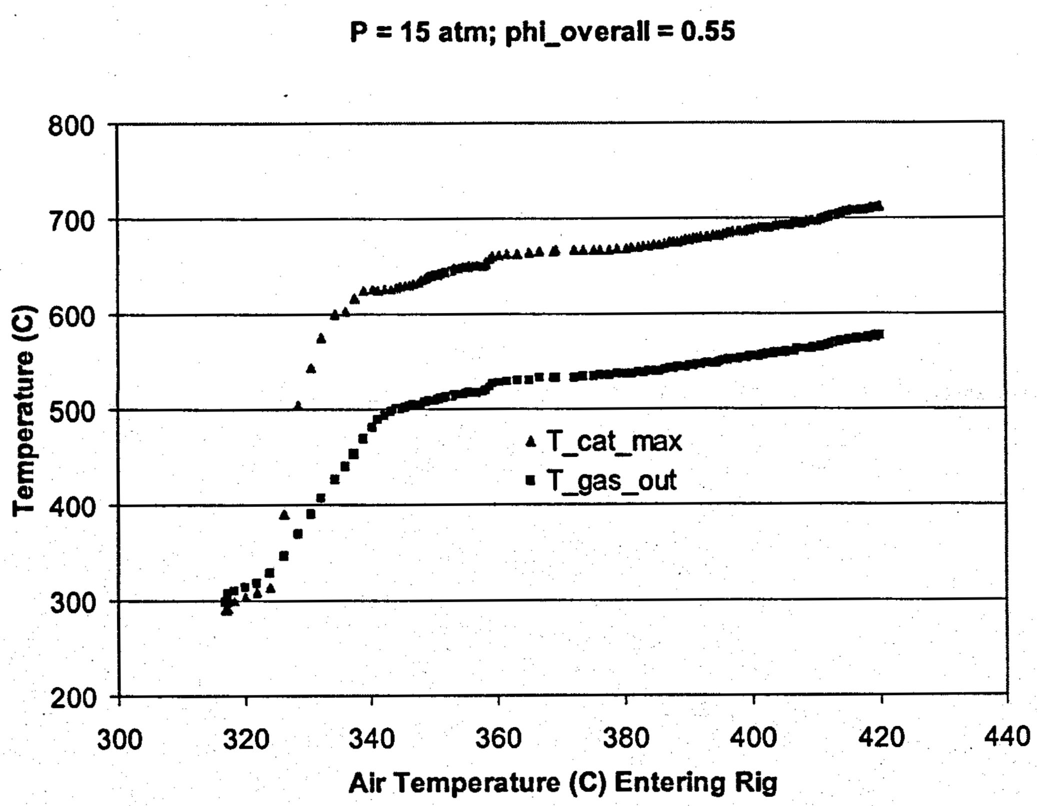

Figure 5.5.7. Maximum catalyst surface temperature ("T_cat_max") and gas temperature exiting catalyst module ("T_gas_out_3") versus rig air inlet temperature, showing catalyst activation (lightoff) at approximately 600 F (320 C).

Figure 5.5.8 shows steady-state operating temperatures at 15-16 atm pressure, including both catalyst surface temperature and gas temperature near the post-catalyst duct exit, plotted against adiabatic flame temperature at the post-catalyst duct exit (prior to addition of any leakage or cooling air, and prior to gas-phase burnout). Adiabatic flame temperature is calculated for San Diego natural gas (approximately 96% methane and 2% higher-order hydrocarbons), for each estimated equivalence ratio in the post-catalyst duct. Air inlet temperature is taken to be 810 F (430 C), and fuel inlet temperature is assumed to be 70 F (20 C). The estimated equivalence ratio is based on GC measurement in the post-catalyst duct (note that the GC measures methane, but not higher-order hydrocarbons), and the assumption that the post-catalyst duct GC measurements were 10% high in fuel. This assumption is based on post-catalyst duct mixing surveys in PCI's atmospheric-pressure lab, which indicated that the post-catalyst duct gas sample port was located in a 10% fuel-rich region.

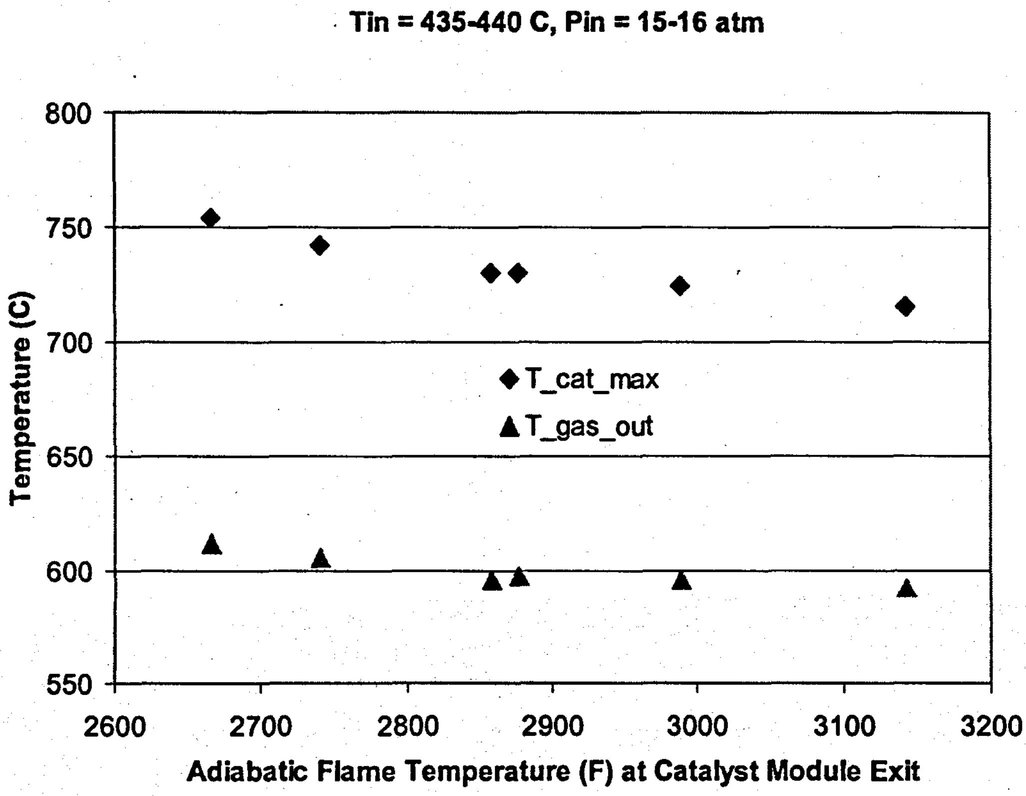

Figure 5.5.8. Maximum catalyst surface temperature ("T_cat_max") and catalyst module gas exit temperature ("T_gas_out") versus rig adiabatic flame temperature at the catalyst module exit.

As shown in Figure 5.5.8, the catalyst surface temperature is fairly insensitive to operating condition, and remains at a fairly constant value, below 780 C (1430 F), over the complete range of operating conditions tested. Likewise, thermocouple-measured gas exit temperatures, at the exit of the post-catalyst duct (but prior to gas-phase burnout) are also insensitive to operating condition, and remain near 600 C (1110 F) over the range of operating conditions tested.

Analysis of the post-catalyst duct samples, however, indicated that the gas temperature at the post-catalyst duct exit should have been approximately 650 - 660 C, based on the heat released as a result of chemical reaction in the catalyst bed. Thermocouple-measured values were probably lower as a result of thermocouple heat loss (error), as well as possible heat loss from the post-catalyst duct.

Combustor Emissions Performance

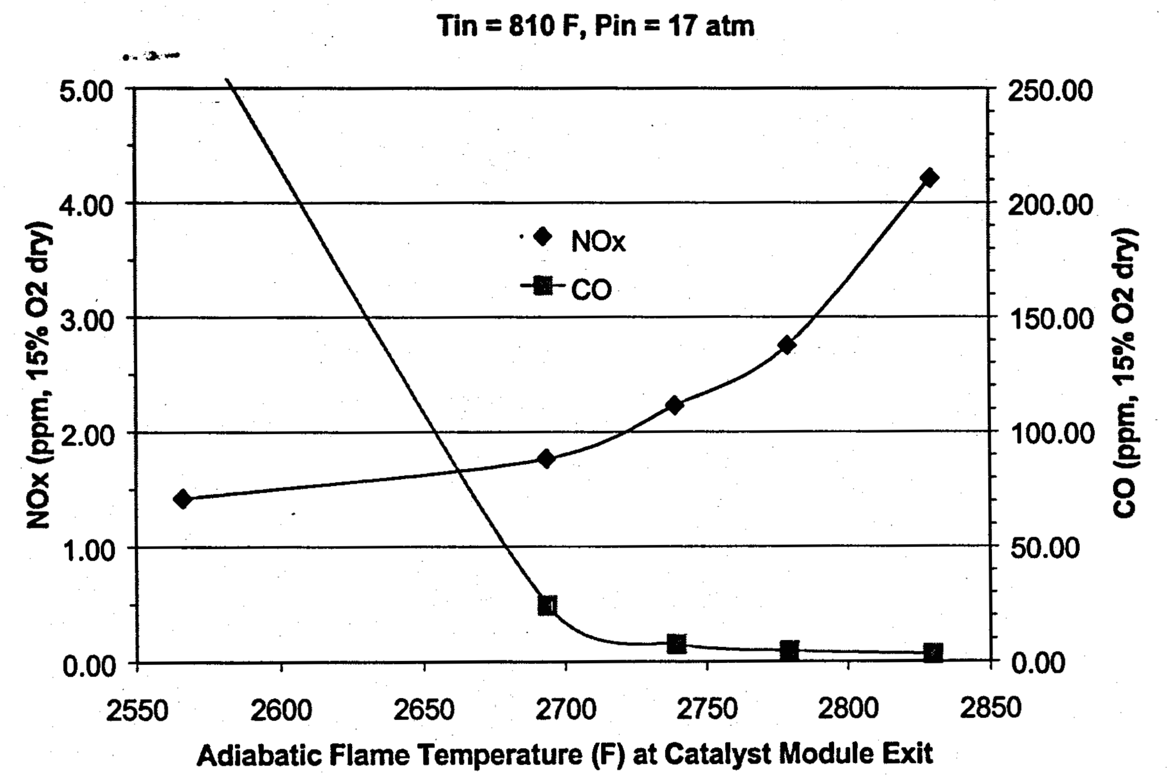

***Figure 5.5.9.** NOx and CO emissions versus adiabatic flame temperature at catalyst module exit, for case with approximately 600 C measured gas temperature exiting the RCL module.*

Figure 5.5.9 shows NOx and CO emissions data (corrected to 15% O_2) over a range of adiabatic flame temperatures from approximately 2550 to 2850 F, for the same RCL module configuration (with 600 C thermocouple-measured post-mix gas temperature) for which data are shown in Figures 5.5.7 and 5.5.8. These data were obtained at 17 atm pressure and 810 F inlet air temperature, with nominally 70 F inlet fuel temperature (natural gas). (The 16-17 atm pressure condition at 810 F inlet air temperature were chosen to be representative of an Advanced industrial gas turbine, such as the Solar Taurus 70.) Air flow to the RCL module was approximately 3 pps. NOx emissions were below 5 ppm at all data points obtained, and were below 3 ppm for flame temperatures less than approximately 2800 F, as expected based on well-mixed lean-premixed combustion. CO emissions were below 10 ppm for flame temperatures greater than approximately 2750 F, but increased to 24 ppm at 2700 F flame temperature, and increased sharply at still lower flame temperatures. UHC emissions were below 5 ppm for flame temperatures greater than approximately 2700 F (not shown). PCI's emissions target of NOx < 3 ppm with CO < 10 ppm was achieved over a flame temperature range of approximately 50 F (data points at 2740 and 2780 F adiabatic flame temperature from module) for this module configuration.

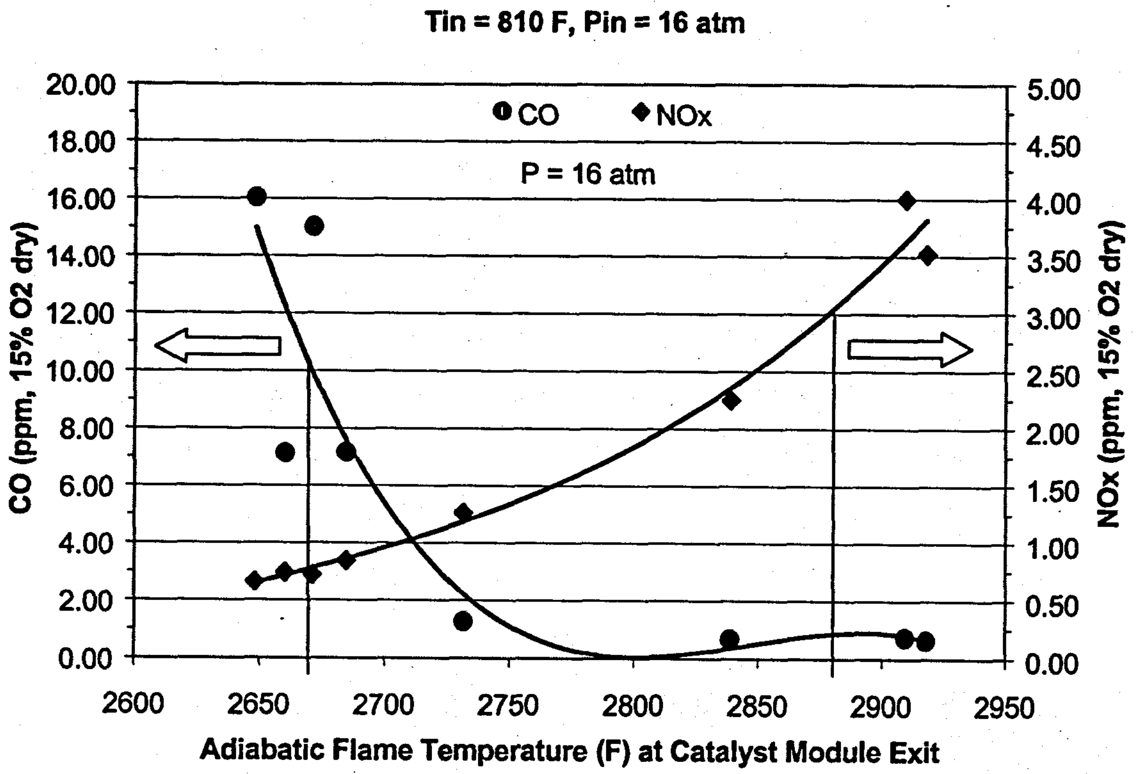

Figure 5.5.10. NOx and CO emissions, as a function of adiabatic flame temperature exiting catalyst module, for case with approximately 650 C measured gas temperature exiting the RCL module. Note approximate 200 F operating window for low emissions (NOx < 3 ppm and CO < 10 ppm).

By further reactor modification, a wider turndown range with ultra-low emissions (NOx < 3 ppm with CO < 10 ppm) was achieved by increasing the gas temperature exiting the catalyst module. For these tests, the thermocouple-measured temperature in the gas stream exiting the post-catalyst mixing duct was approximately 650 C. The data for this case are shown in Figure 5.5.10, at 16 atm pressure (rig operating pressure shifted slightly, from 17 to 16 atm between the tests corresponding to Figure 5.5.9 and 5.5.10). Inlet air temperature was 810 F, and natural gas fuel inlet temperature was again nominally 70 F. As shown, a much improved turndown range of approximately 200 F was obtained with NOx < 3 ppm and CO < 10 ppm (both dry, corrected to 15% O2) by increasing the gas temperature exiting the catalyst.

For this case, analysis of the post-catalyst duct samples again indicated that, based on the heat released as a result of chemical reaction in the catalyst bed, the gas temperature at the post-catalyst duct exit should have been higher than the thermocouple-measured values. For the data shown in Figure 5.5.10, analysis indicates that the zero-heat-loss gas temperature should have been about 690 - 700 C, depending on exact operating condition. Again, thermocouple-measured values were probably lower as a result of thermocouple heat loss (error), as well as possible heat loss from the post-catalyst duct.

Combustion System Pressure Loss

For the data shown in Figure 5.5.9 pressure drop across the combustion system (from shell plenum to combustor exit) was less than 3.75% of shell pressure for all conditions, meeting the Advanced engine requirements.

Combustion-Driven Pressure Oscillations (CDPO, noise, dynamics)

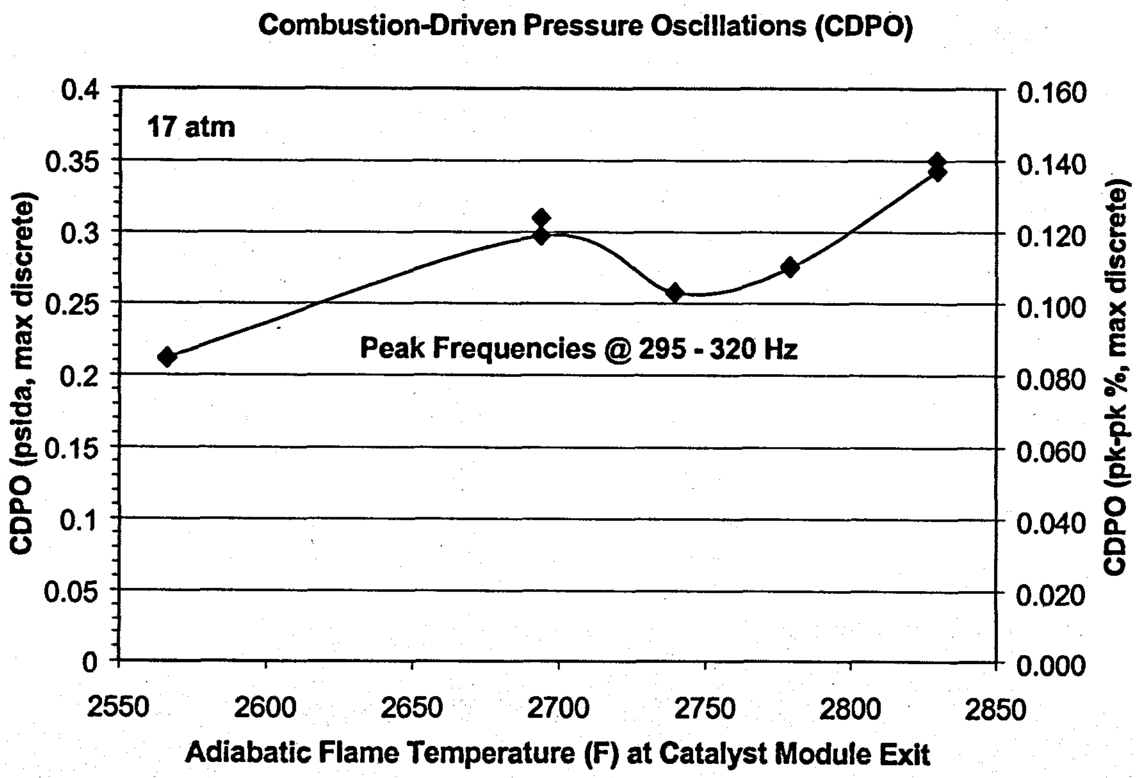

Figure 5.5.11. Combustion-Driven Pressure Oscillations (CDPO) as a function of adiabatic flame temperature at the catalyst module exit, for the same conditions as the Figure 5.5.9 data.

Combustion-driven pressure oscillations (CDPO, "noise", or "dynamics") were insignificant during all tests of PCI's RCL module. The peak noise level, plotted in terms of psida (pounds per square inch double amplitude, or peak-to-peak) at the discrete frequency of maximum amplitude, is plotted in Figure 5.5.11 as a function of adiabatic flame temperature at the module exit. As shown, combustor operation was quiet at all conditions, with pressure oscillations always less than 0.35 psida for this data set (same operating conditions as the data in Figure

5.5.9). In terms of percent shell pressure, peak-to-peak noise was less than 0.14% at all conditions (shell pressure was 17 atm for this data).

Combustion noise data were also obtained for the conditions of Figure 5.5.10, and were again insignificant, with peak noise levels again always less than 0.35 psida.

6. CONCLUSIONS / NEXT STEPS

Under this program, the RCL concept was successfully developed to address the need of Advanced industrial engines to operate with low single-digit NOx emissions. Ultra-low emissions (NOx < 3 ppm with CO < 10 ppm) were achieved in full-scale high-pressure tests at Solar Turbines, in a catalytic combustion system offering the following additional benefits: operation without a pre-burner, and with relaxed unmixedness requirements; robust operation; simple single-fuel control system; and compact size for engine integration and retrofit applications.

Based on the successful full-scale combustion tests and sub-scale durability tests, the program met its stated objectives of achieving NOx < 5 ppm and technical potential for 8000 hour catalyst durability. In addition, cost modeling indicates that in volume production RCL reactor cost will be less than 15% of machine cost.

For Advanced industrial engine applications, the next step will be catalytic combustor testing in an engine rig at Solar Turbines. Fabrication of four (4) RCL modules is underway for this purpose, with engine testing currently scheduled for Spring 2002. Following a successful outcome of this testing, a design task should be initiated for RCL implementation in a production industrial engine, such as Solar's Taurus 70.

7. REFERENCES

D.N. Anderson, R.R. Tacina, and T.S. Mroz (1975). NASA Technical Memorandum X-71747.

Dalla Betta, R.A., Schlatter, J.C., Nickolas, S.G., Cutrone, M.B., Beebe, K.W., Furuse, Y., and Tsuchiya, T. (1997). *Journal of Engineering for Gas Turbines and Power*, **119**, pp. 844-851.

P. Dutta, D.K. Yee, and R.A. Dalla Betta (1997). ASME Paper No. 97-GT-497.

J.P. Kesselring, W.V. Krill, E.K. Chu, and R.M. Kendall (1979). In proceedings of **New fuels and advances in combustion technologies** symposium, Mar. 26-30, 1979, New Orleans, LA.

W.C. Pfefferle (1974). U.S. Patent No. 3,846,979.

Pfefferle W. P., Etemad S., and Karim H., (June, 1996). Advanced Technology Catalytic Combustor for Natural Gas Tubine Engines, *Final Report, Phase 1*, DOE # DE-FG02-95ER82054.

Pfefferle, W.C. (1997). U.S. Patent No. 5,601,426.

P.W. Pillsbury (1984). ASME Paper No. 84-GT-54.

T.J. Rosfjord (1976). AIAA Paper No. 76-46, Washington DC, Jan. 1976.

D.A. Smith, S.F. Frey, D.M. Stansel, & M.K. Razdan (1997). ASME Paper No. 97-GT-311.

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