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Advanced technology catalytic combustor for high temperature ground power gas turbine applications

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Abstract

We report results from a lean burn ultra-low emission catalytic combustor. In a sub-scale rig, atmospheric testing with methane demonstrated $NO_x<3$, CO<5, and UHC<1 ppm, with stable combustion at inlet temperatures of 400–500°C (750–1020°F) and combustor discharge temperatures of 1150–1540°C (2100–2800°F). Catalyst temperatures were held well below metal substrate material limits, while combustor discharge temperatures of up to 1540°C (2800°F) were achieved. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Worldwide demand for ground power generation gas turbine engines is burgeoning, while improved technology is allowing significantly higher efficiency designs. Yet tightening emissions regulations are being applied worldwide to the gas turbine combustor ground power generation market. Dry low NO_x combustion technologies now under development use high levels of aerodynamic stabilization and combustor staging to allow lean premixed prevaporized (LPP) operation at adiabatic flame temperatures where thermal NO_x formation is substantially reduced. To keep NO_x below 15–25 ppm, these designs must operate very close to their stability limits, severely limiting low emissions turndown and increasing the problem of combustion acoustic instabilities. A diffusion flame stage or pilot burner is generally used for start-up and for operation below stable premix conditions. As a result, designs are complex with multiple stages, at part-load conditions diffusion stage operation causes NO_x formation, and combustion instabilities remain a problem. Catalytic combustor designs can overcome many of these problems, providing a potentially superior design if durability and operational issues are resolved.

Currently, the best available control technologies for NO_x reduction require exhaust gas after-treatment. Selective catalytic reduction (SCR) after-treatment of gas turbine exhaust is capable of achieving low single digit ppm NO_x emissions (corrected to 15% O_2), with a similar amount of ammonia slip (which is functionally equivalent to NO_x). SCR has both high capital costs and ongoing operating costs, impairing cost-effectiveness and efficiency. These combined costs have dropped in recent years, but remain in the approximately 15–20% range of the purchase price of the engine. Catalytic combustion offers to reduce NO_x emissions from current targets of 9–15 ppm to the

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1–2 ppm range, i.e. even lower than is generally feasible with SCR.

Catalytic combustion offers high efficiency and emissions sufficiently low that post combustion treatment is not needed [12,13]. It is well established and repeatedly documented that catalytic combustion can achieve stable combustion of premixed natural gas and air at low enough temperatures that NO_x emissions are insignificant (e.g. 1 ppm or so), even at pressure conditions [2,8]. Unburned hydrocarbons and CO emissions can simultaneously be held very low (e.g. single digit ppm) by operating well within the stability limit yet well below the temperature at which thermal NO_x production is significant. Catalytic systems can also have the benefit of reduced combustion instability/acoustics, which is a major issue with dry low NO_x engines. In addition, catalytic systems have been developed for use with liquid fuels as well. In summary, catalytic combustion systems have the following advantages:

- Leaner combustion.
- Lower peak flame temperatures.
- Lower emissions: NO_x, CO, UHC.
- Reduced combustion instability.

Although catalytic combustion has been shown to be beneficial, commercial catalytic combustor development has been restrained by the following [5–8]:

1. Catalyst and support materials durability. No catalyst system has previously been demonstrated to operate reliably at the turbine inlet temperatures dictated by ATS operating conditions.
2. Fuel/air preheating required for sustaining catalytic reactivity given available compressor discharge temperatures (generally requiring a preburner, with resulting NO_x emissions no longer significantly below those of dry low NO_x pilot burners).
3. Turndown limits due to a relatively narrow operating band between minimum required inlet temperature (surface kinetics limited) and maximum survivable catalyst temperature.
4. Uniform fuel/air mixture is required to avoid catalyst substrate meltdown as a result of localized rich zone operation.
5. High pressure drop associated with conventional catalytic combustion systems, since a long monolith is required to convert all of the fuel/air mixture to products.

2. Objectives

The purpose of the present study is to demonstrate the potential of a catalytic combustor for high temperature modern ground power gas turbine engine applications. Consistent with the Department of Energy's (DOE) Advanced Turbine System (ATS) main program goals [3], the objectives are:

- *Efficiency*: 15% improvement in efficiency as compared to the best 1991 technology in each class. In general, an increased firing temperature is sought to increase the performance level.
- *Emissions*: 10% reduction in emission levels, with UHC and CO emissions below 20 ppm and NO_x emissions below 10 ppm. The higher firing temperatures required to increase efficiency make NO_x reduction particularly challenging.
- *Durability*: Similar durability to present state-of-the-art designs. This requires new designs to compensate for higher firing temperatures, while delivering the same engine lifetime as current designs.

Several engine manufacturers have initiated cycle plans to meet these requirements [1]. In addition to cycle plan, typical operating parameters for advanced, high-temperature, high-efficiency gas turbine engines were described [1], which motivated the operating conditions used for the present study, as shown below:

1. Combustor inlet temperature $T_{3.1}$ above 400°C (930°F). A range of temperatures between 400°C and 500°C (750°F and 930°F) was used for the present study.
2. Combustor exit temperature $T_{3.8}$ below 1540°C (2800°F). Higher temperatures were also explored in the present study.
3. Inlet velocity over the catalytic reactor in the range 15–30 m/s (50–100 fps), covering a wide range of engine operation.

3. Technical approach

In this study the catalytic combustor technology integrates catalytic reaction zones and downstream aerodynamic flame stabilization to achieve effective, durable catalyst operation and to extend the lean stability limit for natural gas applications. Based on our previous work on catalytic combustion of liquid fuel, we observe that methane combustion requires higher catalytic activity and better downstream design due to the higher stability of methane. Following are some of the features implemented in the present approach to achieve a practical design directed towards natural gas combustion.

3.1. *Catalytic reaction zone*

High palladium content monolithic catalysts are placed upstream the aerodynamic flame zone to sustain ATS cycle operation from start-up to idle to part load without use of a preburner. Short length catalytic monolith were chosen to ensure that the catalyst temperature remains well below the adiabatic flame temperature. The catalytic surfaces are kept cool enough so that the catalyst substrate can be fabricated out of high-temperature metals. The target temperature range for the catalyst is below 900°C (1650°F). As a result the combustor life is improved and the catalyst reactor section is not a life limiting component.

3.2. *Aerodynamic flame stabilization zone*

An aerodynamic flame zone just downstream of the catalytic reactor provides a low velocity region where heat and reactive products of incomplete combustion from the catalysts are leveraged to achieve ignition and a core of flame stabilization in conditions beyond the normal lean limits. Primary heat release is thus achieved downstream of the catalytic reactor. The use of active aerodynamic flame stabilization to continuously ignite and stabilize combustion of the preactivated fuel/air mixture limits the proportion of total reaction required in the catalytic zone.

4. Experimental

4.1. *Test rig fabrication*

A versatile test rig shown in Fig. 1 was constructed for this study. The rig was modular in construction and consisted of a ceramic lined section, housing the catalyst monoliths, and a separate downstream combustor housing. The two sections were joined at the gas-phase flame stabilization location. The modular construction allowed rapid rebuilds and configuration changes. A quartz window was included to view the catalytic reactor region during testing.

A blower was used to supply air. Air velocity was measured with a digital rotary vane meter. An electric heater section was fabricated and used for air preheating. Inlet gas temperatures were monitored with cold referenced type K thermocouples. A fuel nozzle and static mixer system were used for methane injection. A mass flow controller and digital readout were used to precisely control and measure the methane flow. The combustor front section was bolted onto the end of the diffuser and could be separated easily to allow the prototype to be readily accessed.

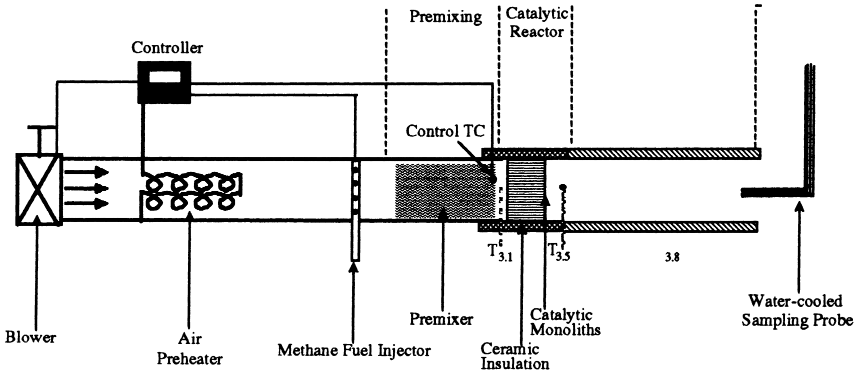

Fig. 1. Schematic of test rig assembly.

The catalyst housing was constructed with a removable top plate which allowed easy access to the internal area, and was flanged to reduce leakage. The internal surfaces were lined with 12.5 cm ($\frac{1}{2}$ in.) thick ceramic insulation. The combustor housing was designed to allow gas sampling and extraction. Type S thermocouples were used to monitor internal temperatures within the catalytic combustor and at the combustor exit. Exhaust gas exited the rig and was removed by a large exhaust blower mounted on the roof of the facility.

4.2. Emissions measurement

Exhaust emissions including UHC, CO, CO_2, O_2 and NO_x were measured at a number of different steady state conditions. A water cooled rake type sampling probe was designed and built to provide six zones of equal area exhaust gas sampling. The rake was located 0.4 m (1.3 ft) downstream of the catalytic reactor exit, providing a gas residence time of 50–100 ms from the catalytic reactor exit to the rake type sampling probe for the range of velocities tested. A sample delivery pump, calibrated for the desired amount of sample gas flow, was used to draw the exhaust sample through the sampling probe and into the analytical instruments for emissions detection.

The emissions system used for the testing consists of a flame ionization detector based total unburned hydrocarbon (UHC) analyzer, nondispersive infrared detector for carbon monoxide (CO), a nondispersive infrared carbon dioxide (CO_2) detector, a paramagnetic oxygen (O_2) detector, and a chemiluminescent analyzer for oxides of nitrogen (NO_x). Each instrument was mounted in a fully portable rack, with pumps and back-pressure valves to maintain constant pressure and flow to all instruments. A thermoelectric chiller was used behind the UHC analyzer to cool (to 4°C) the heated sample gas and condense out the water before delivery to the other instruments. The instruments were zeroed and spanned with bottled calibration gases before each test series to assure accurate and precise measurements. Data were obtained directly from the instruments and recorded.

4.3. Test conditions

A series of tests was performed. Aside from design choices, test variables included burner inlet temperature, combustor outlet temperature, fuel/air ratio, and inlet velocity. For each steady-state test condition tested, measurements were made of NO_x, CO, and UHC emissions, fuel flow, air flow, and temperatures at several locations, including air inlet, combustor outlet, and catalyst reactor exit. Selecting three suitable baseline inlet temperatures of 400°C, 450°C, and 500°C (750°F, 840°F, and 930°F), and inlet velocities in the range 12–30 m/s (40–100 fps) where stable combustion could be maintained, the fuel–air ratio was varied to determine maximum combustor outlet temperatures for durable operation from 1100°C (2000°F) up to at least 1540°C (2800°F). Higher outlet temperatures, as high as 1775°C (3225°F), were also explored. Inlet velocities were explored as a variable across the range from 12–30 m/s (40–100 fps). The catalyst surface was examined for durability at the end of each experiment set. For the same three inlet temperatures, the lean limit of operation was established by lowering the fuel flow to the point of notable increase in carbon monoxide emissions and/or flame extinction. Pressure drop was measured for the final prototype across a range of velocities from 12 to 30 m/s (40–100 fps) for the three inlet temperatures of 400°C, 450°C, and 500°C (750°F, 840°F, and 930°F).

5. Discussion of results

For the present work, catalysts were evaluated for light-off below 400°C (ATS condition), for sufficient conversion to sustain the downstream flame without overheating the catalyst reactor, and for catalyst durability. Fig. 2 shows typical data for the catalyst used during the present work. This figure shows that the inlet temperature increases to approximately 330°C, where catalyst light-off occurs and the exit temperature of the catalyst reactor zone increases due to partial fuel conversion. At about 400°C, a temperature increase of 80°C is achieved. At 500°C a temperature increase of 100–170°C was obtained across the catalyst reactor zone, depending on stoichiometry. The conversion rate increased at equivalence ratios nearer to stoichiometric. However, for the range of stoichiometries studied the temperature increase across the catalytic reactor was never more than 200°C. This is important for avoiding catalyst overheating and assuring catalyst durability. Light-off temperature was

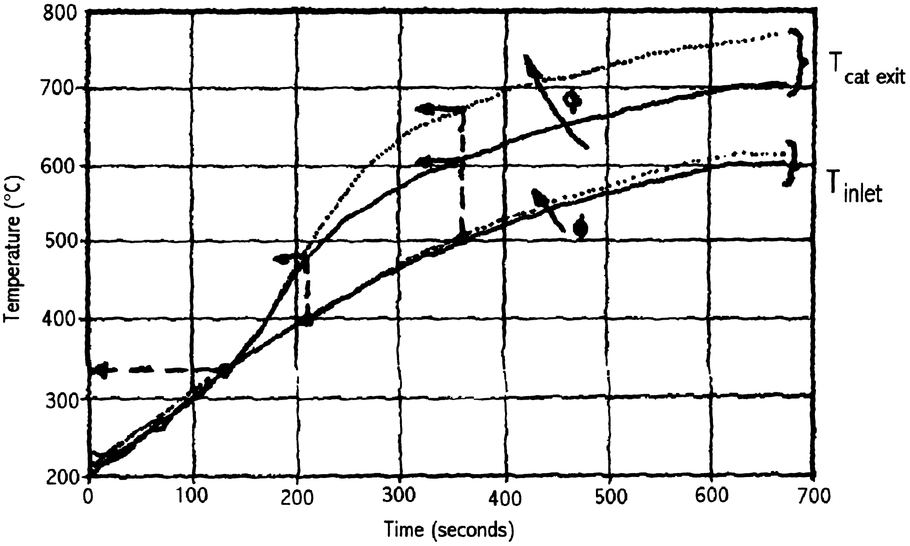

Fig. 2. Typical catalyst formulation performance demonstration.

studied for a range of velocities and, as expected, was found to be independent of velocity. The catalyst coating used for this work was found to be capable of light-off below 400°C and provided adequate conversion such that the flame downstream of the reactor was sustained without overheating of the catalyst bed region. Overall, the catalyst coating used for this study was demonstrated to be effective throughout the cycle range.

Pollutant emissions and catalyst bed exit temperature are shown in Fig. 3 as functions of combustor exit temperature for simulated ATS conditions (400°C (750°F) inlet temperature, 21 m/s (70 fps) inlet velocity). Combustor exit temperature was varied by adjusting the premixed fuel–air ratio. All emission data shown in this and subsequent figures are corrected to 15% O_2 concentration. As expected, the concentration of oxides of nitrogen (NO_x) increased with increasing combustor exit temperature, due to a greater rate of thermal NO_x production at higher temperatures. Overall, NO_x concentrations remained below 3 ppm across a wide range of combustor outlet temperatures (1200–1600°C). Note that typical gas residence times in gas turbine engine combustors are 10–20 ms, compared to a gas residence time of 50–100 ms between the exit of the prototype catalytic reactor and quenching of reactions in the rake type sampling probe. Since thermal NO_x concentrations increase with greater residence times [11], the NO_x measurements we report here can be expected to be less at lower residence times. In addition, because NO_x concentrations are largely independent of pressure for lean premixed combustion [4,14], the results reported here are indicative of expected NO_x emissions at higher pressure conditions, assuming same level of mixedness.

Total unburned hydrocarbon (UHC) concentration is shown in Fig. 3 to decrease with increasing combustor exit temperature. Again, this is an expected result since UHC emissions generally decrease at higher temperatures due to improved fuel dissociation and increased reaction rates [9]. UHC emission from the catalytic combustor prototype was always very low, and never exceeded 0.5 ppm. At actual engine conditions even lower UHC emission is expected, due to increased chemical reaction rates. Low UHC emission indicates full conversion of fuel into products, with a resulting potential for high engine efficiency.

For temperatures between 1200°C and 1300°C, CO concentration decreased with increasing combustor exit temperature, reaching a minimum at about 1300°C. At temperatures below 1300°C local flame extinction and slow chemical reaction rates cause CO emissions to be greater than the minimum value. At temperatures greater than 1300°C CO concentration increased with increasing combustor exit temperature as a result of chemical dissociation at higher tempera-

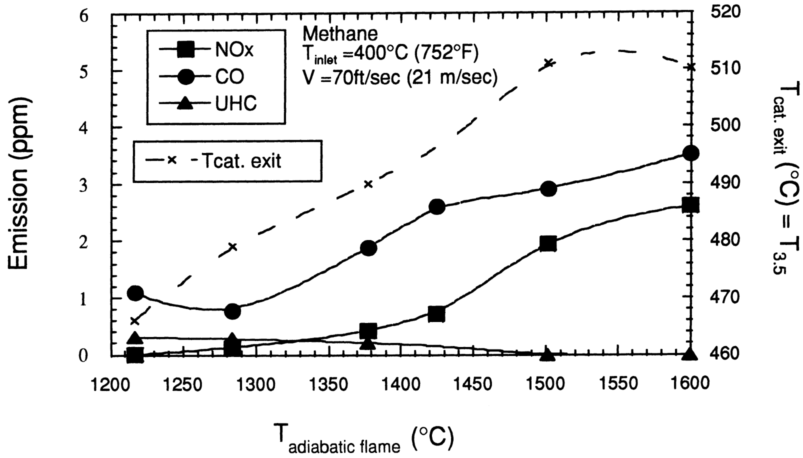

Fig. 3. Pollutant concentrations (NO_x, CO, and UHC) in exhaust gas at different adiabatic flame temperatures. Catalytic reactor exit temperatures are also shown.

tures (greater equilibrium CO concentrations at higher temperatures) [10]. All the CO data obtained show this trend, regardless of operating condition. In all cases (all temperatures, all operating conditions) the measured CO concentration was less than 5 ppm. Higher combustion pressures would reduce CO concentrations by increasing the rate of CO conversion to CO_2, and by suppressing dissociation of CO_2 at high temperatures [10].

Also shown in Fig. 3 is the catalyst bed exit gas temperature (dotted line) which was far below the metal material limit (900°C (1650°F)) over the entire turndown range from approximately 1200°C to 1600°C at the combustor exit, showing the potential for high durability of the present catalytic combustor. While the actual surface temperature may be expected to climb above the gas temperature, the margin of safety is sufficiently wide that durability may be inferred.

Catalyst reactor exit gas temperature and combustor exit gas temperature are shown in Fig. 4 as functions of inlet temperature, for reactant velocity of 18 m/s (60 fps). For an inlet temperature range 400–500°C the catalyst reactor temperature remained below the

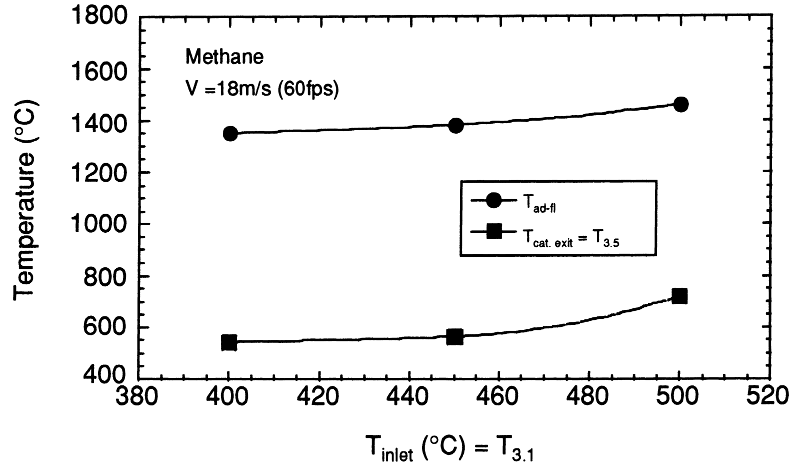

Fig. 4. Catalytic reactor exit temperature and adiabatic flame temperature at different inlet temperatures.

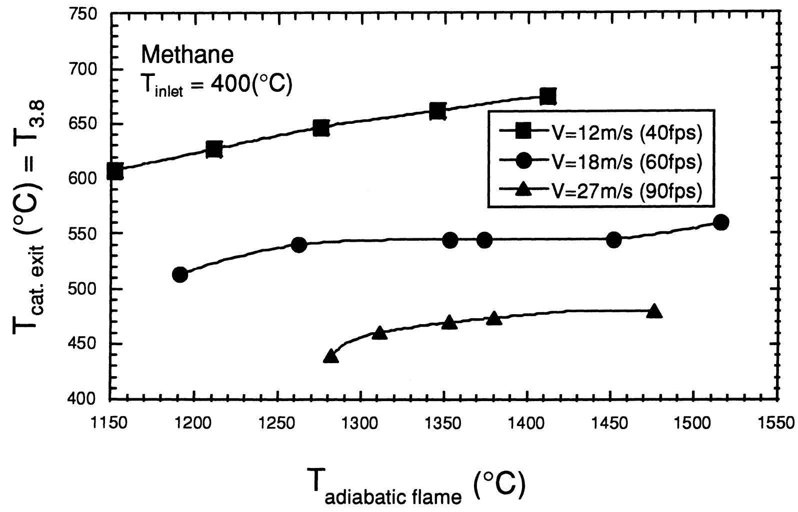

Fig. 5. Catalytic reactor exit temperature for different adiabatic flame temperatures and inlet velocities of 12, 18, and 27 m/s.

gas-phase ignition temperature, and well below the metal material limit of 900°C (1650°F). For all testing conducted, up to combustor temperatures of 1770°C (3220°F), the reactor exit temperatures were always measured to be below 675°C (1250°F). These low temperatures permit safe use of metal substrates, overcoming the need and cost of ceramics development, and providing higher durability.

Fig. 5 shows the effect of inlet velocity (gas residence time) on catalyst reactor exit gas temperature. It is observed from the figure that an increased inlet velocity requires an increased equivalence ratio to maintain a stable gas-phase flame downstream of the catalytic reactor. The figure also shows that the present catalytic combustor type arrangement permits safe temperatures for metal substrates over a wide range of velocities. A decrease in air flow through the combustor (up to a factor of 2), which may occur during compressor surge, will not damage the catalyst bed. This is an important factor in terms of engine transient operation. These data indicate that the catalytic reactor section temperature range is similar to existing engine front ends (e.g. swirler, header, etc.) and as a result similar durability in existing engine applications is expected, thus meeting the ATS goal of durability.

Figs. 6 and 7 present corrected exhaust gas NO_x concentrations for the two extreme inlet temperatures (400°C and 500°C, respectively) as a function of combustor exit temperature at several different inlet velocities. Figs. 8 and 9 show that lean low temperature combustion, made possible by premixing and catalytic prereaction, limits NO_x emissions to below 3 ppm at all operating conditions, well below the ATS goal of NO_x<10 ppm. Catalytic prereaction enables stable combustion at low equivalence ratios, and the resulting low adiabatic flame temperatures inhibit the formation of thermal NO_x [11]. The figures also demonstrate the adverse effect of increased gas residence time in the combustor, as indicated by increased NO_x formation at lower reactant inlet velocities. As discussed earlier, the 50–100 ms residence time between the catalytic reactor exit and the exhaust gas sampling probe is long compared to typical gas turbine engine combustor residence times of 10–20 ms. Thus, NO_x emissions from the present catalytic combustor in actual engine conditions are expected to be less than reported here.

Corrected exhaust gas CO concentrations are shown in Fig. 8 as a function of combustor exit temperature for three different inlet velocities. UHC emissions are not plotted here since at all conditions UHC emissions were below 1 ppm. Both CO and UHC emissions are well below the ATS maximum allowed value of 20 ppm. Note that inlet velocity (residence time) has a minimal effect on CO emissions. At high-pressure conditions CO emissions are expected to be still lower than shown, due to improved combustion and less dissociation of CO_2. Wide turndown capability at low emissions is observed for all the figures.

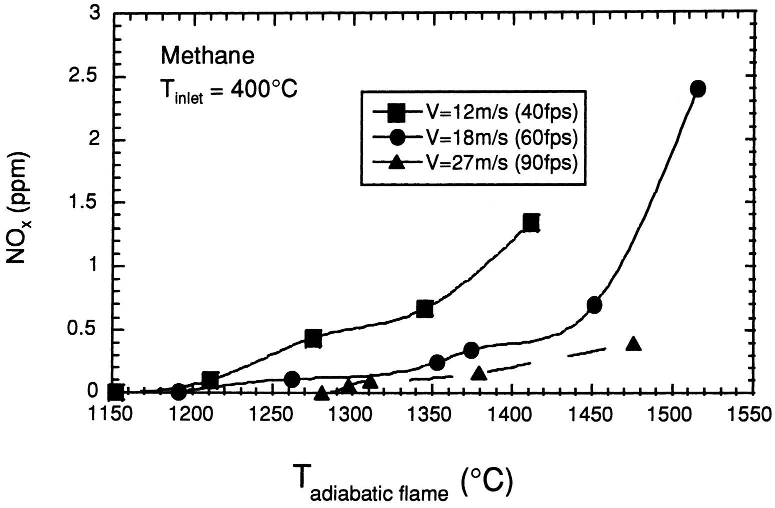

Fig. 6. Measured NO_x emissions at different adiabatic flame temperatures (for an inlet temperature of 400°C).

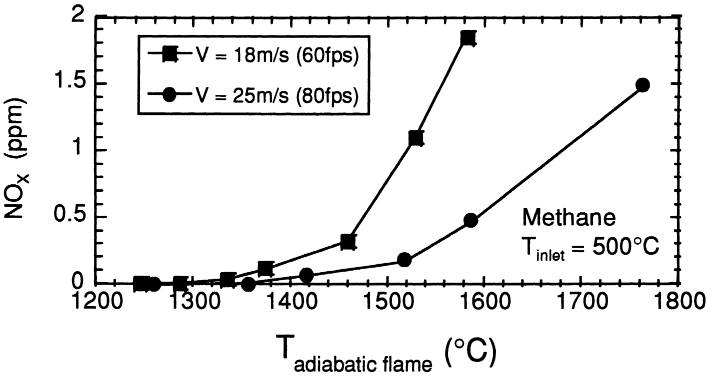

Fig. 7. Measured NO_x emissions at different combustor exit temperatures, for an inlet temperature of 500°C.

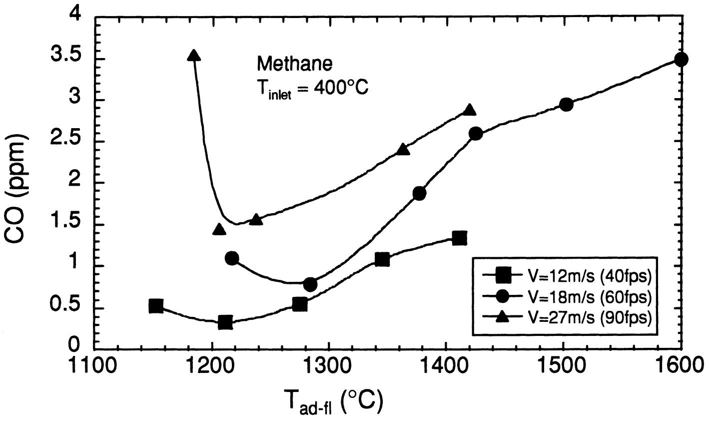

Fig. 8. Measured CO emissions at different adiabatic flame temperatures for an inlet temperature of 400°C.

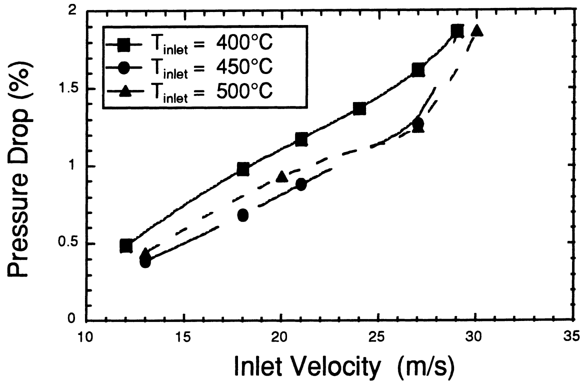

Fig. 9. Percent pressure drop across ATCC combustor for different inlet temperatures and different inlet velocities.

Percent pressure drop, as shown in Fig. 9 for different inlet velocities and temperatures, is below 2%. This low level of pressure drop will assure an improved engine efficiency, targeted by DOE ATS requirements of 60%. The measured pressure drop was across the entire catalytic combustor device (catalytic reactor and downstream aerodynamic sections); future optimization of the downstream aerodynamic section will further reduce the level of pressure drop. The upstream premixer was not included in the pressure drop measurement, since it was outside the scope of the present study, and is specific to engine configuration. It should be noted that the present design, with resulting low catalyst surface temperatures, is less sensitive to premixing non-uniformity than conventional catalytic combustors, simplifying the design of a low-pressure drop premixer.

6. Conclusions

The present work demonstrated successful implementation of the catalytic combustor concept in a subscale rig operating at simulated ATS conditions. In this study we illustrated the ability to sustain combustion at very low local equivalence ratios and to provide turndown capability with low emission and without overheating the metal monolith. Operations at flame temperatures over the range from 1200°C (2200°F) to 1540°C (2800°F) were demonstrated in this study. The results show that the design has potential for meeting all the DOE ATS requirements by a safe margin. Proper implementation in a ground power gas turbine engine should assure similar positive results.

Decoupling of the catalytic reactor zone from the gas-phase zone made the design durable over a wide range of operating conditions, thus demonstrating its suitability for modern, high-temperature, high-efficiency engines. Furthermore, the low combustor pressure drop of the design will assure that engine manufacturers meet ATS efficiency goals.

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