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EFFECT OF INLET TEMPERATURE ON THE PERFORMANCE OF A CATALYTIC REACTOR

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NOTICE

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ABSTRACT

A 12-cm diameter by 15-cm long catalytic reactor was tested with No. 2 diesel fuel in a combustion test rig at inlet temperatures of 700, 800, 900, and 1000 K. Other test conditions included pressures of 3 and 6×10^5 Pa, reference velocities of 10, 15, and 20 m/s, and adiabatic combustion temperatures in the range 1100 to 1400 K. The combustion efficiency was calculated from measurements of carbon monoxide and unburned hydrocarbon emissions. Nitrogen oxide emissions and reactor pressure drop were also measured. At a reference velocity of 10 m/s, the CO and unburned hydrocarbons emissions, and, therefore, the combustion efficiency, were independent of inlet temperature. At an inlet temperature of 1000 K, they were independent of reference velocity. Nitrogen oxides emissions resulted from conversion of the small amount (135 ppm) of fuel-bound nitrogen in the fuel. Up to 90 percent conversion was observed with no apparent effect of any of the test variables. For typical gas turbine operating conditions, all three pollutants were below levels which would permit the most stringent proposed automotive emissions standards to be met. The pressure drop increased linearly with reference velocity and decreased slightly as the inlet temperature was raised. Pressure drop increased linearly with velocity to a maximum value of 1.5 percent at a reference velocity of 20 m/s.

INTRODUCTION

Probably the most important aspect of catalytic combustion is the production of extremely low concentrations of thermal NO_x. Because of this feature, evaluations are being made of catalytic combustion for aircraft (refs. 1 to 3), stationary (refs. 4 to 6), and automotive applications.

Catalytic combustion is being studied at the NASA-Lewis Research Center for the Gas Turbine Highway Vehicle Systems Project which is supported by the Department of Energy. The goal of this project is to demonstrate the technology for an automotive gas turbine engine which would improve the thermal efficiency over current spark-ignition piston engines. This improved gas turbine is presently being defined by each of the four major American automobile manufacturers under development contracts to the DOE. The engine is expected to operate with a turbine inlet temperature of about 1300 K and with combustor inlet temperatures which will decrease from 1200 K at idle to 1000 K at full speed (ref. 7).

Combustor work in support of this Project involves studies of fuel-air premixing/prevaporizing systems (refs. 8 and 9), and the evaluation of catalytic reactors (refs. 10 and 11). Monolithic catalytic reactors have been tested at steady-state conditions which simulated those of the improved engine combustor except that the maximum test section inlet temperature was only 800 K. Several studies (refs. 1, 10, and 12) have shown that an increase in inlet temperature results in an improvement in catalyst performance; however, none of those studies reported results for temperatures higher than 800 K.

The present study was made with test section inlet temperatures as high as 1000 K. While this temperature matches the improved gas turbine combustor inlet at full speed, it is 200 K lower than the idle inlet temperature. To provide a basis for extrapolation of results to the idle condition, reactor performance was determined at inlet temperatures of 700, 800, and 900 K as well. Other test conditions included pressures of 3 and 6×10^5 Pa, reference velocities (catalytic reactor inlet velocities) of 10, 15, and 20 m/s, and a range of adiabatic combustion temperatures of 1100 to 1400 K. The catalytic reactor was 12 cm in diameter and 15 cm long. No. 2 diesel fuel was used for all tests.

EXPERIMENTAL DETAILS

The test rig is described in figure 1(a). All ducting was made from 15.2-cm (6-in. nominal) diameter stainless steel pipe with 12 cm ID by 15.2 cm

OD Carborundum T30R Fiberfrax tube insulation inserted inside the pipe to give a flow diameter of 12 cm.

Test section inlet air was indirectly preheated to temperatures in the range of 700 to 1000 K. The inlet temperature was measured before fuel injection at a plane 40 cm upstream of the catalytic reactor. Twelve Chromel-Alumel thermocouples were positioned in this plane at the centers of equal duct cross-sectional areas. In previous studies at Lewis (refs. 7 and 11), the inlet fuel-air mixture temperatures were measured nearer the reactor inlet plane to avoid cooling errors associated with the poorly insulated pipes used in those tests. The difference between the inlet air temperature and the inlet fuel-air mixture temperature depends on the fuel-air ratio, but should be no more than 15 to 20 K for the conditions of these tests.

A multiple conical tube fuel injector was located 15 cm downstream of the inlet thermocouple plane and 25 cm upstream of the catalytic reactor. This fuel injector was of the same type developed by Tacina (refs. 8 and 9); it is pictured in figure 1(b). Two separate sets of 21 equal-length fuel tubes introduced fuel at the small-diameter (high-air-velocity), upstream end of the conical airflow tubes. The large-diameter (1.6 mm) fuel tubes were for propane which was not used in this study, and the small-diameter (0.5 mm) were for the No. 2 diesel fuel used in this study. Tests with this injector type have shown that, 24 cm downstream of the fuel injector inlet, the fuel-air ratio and the velocity varied by less than ±10 percent over the duct cross-section (ref. 8). With an inlet air temperature of 700 K or greater, fuel vaporization approached 100 percent at a distance of 17.8 cm (ref. 9). Therefore, at 25 cm the fuel should be fully vaporized and mixed. The pressure drop reported in reference 13 for this type of fuel injector was about 0.25 percent at a reference velocity of 10 m/s, increasing to 1 percent at 20 m/s.

The reactor inlet pressure was measured at a tap 13 cm upstream of the catalytic reactor. At the same location a single Chromel-Alumel thermocouple was used to detect burning upstream of the reactor. None was observed.

The catalytic reactor was the same J4 reactor used in two earlier studies (refs. 11 and 14). It consisted of two 12-cm diameter and 7.5-cm long metal-foil monolithic elements placed end to end and separated by a 0.31-cm diameter Pt vs Pt-13 percent Rh thermocouple which measured the temperature at the duct centerline. The elements are described in Table I; they were identical except that the first element used a Pt catalyst and the second Pd. Previous to the start of testing this reactor had been operated for about 30 hours at adiabatic combustion temperatures as high as 1500 K.

An array of 12 Pt vs Pt-13 percent Rh thermocouples were used to measure the temperature at the reactor exit plane (see fig. 1(a)). The reactor pressure drop was measured with a differential pressure transducer which indicated the difference between the inlet static pressure and the static pressure at a tap 7 cm downstream of the reactor.

An array of 11 Pt vs Pt-13 percent Rh thermocouples measured the average temperature at a plane 22 cm downstream of the reactor. At the same plane a sample of the exhaust gas was obtained at the centerline of the duct with a single-point probe. The probe was water-cooled to quench the sample near the entrance port. The quenched gases flowed from the probe through an 18-m length of 0.5-cm diameter electrically heated stainless-steel tubing to the gas analyzers. This sample line was maintained at 410 to 450 K to prevent the condensation of any unburned hydrocarbons in the sample. Concentrations of CO and CO_2 were determined with Beckman Model 315B nondispersive infrared analyzers, unburned hydrocabons with a Beckman Model 402 flame ionization detector, and nitrogen oxides (NO and NO_2) with a Thermo Electron Model 10A chemiluminescent analyzer. Before analyzing for CO, CO_2, or NO_x, water vapor was removed with a Hankinson Series E refrigeration-type dryer. Corrections were made to the measured concentrations to obtain the actual, wet-basis concentrations of these three constituents.

After discharging from the downstream instrumentation section, the exhaust gas passed through a water spray to cool the combustion gases, then through a back-pressure valve for control of rig pressure.

MEASUREMENTS AND COMPUTATIONS

For each setting of inlet airflow, temperature, and pressure, data were obtained at several different fuel flowrates, first with fuel flow increasing, then decreasing. This procedure permitted a check to be made of the repeatability of the data, and to determine if hysteresis effects occurred. The repeatability was excellent and no hysteresis was evident.

The reference velocity was computed from the measured mass flowrate, the average temperature measured at the test section inlet plane, the duct cross-sectional area, and the pressure measured at the test section inlet. Thus, the reference velocity is the same as the reactor inlet velocity.

The emissions were measured as concentrations in ppm by volume and converted to emission indexes using the expression

$$(E.I.)_x = C_x \times 10^{-3} \frac{1 + f}{f} \frac{M_x}{M_p}$$

where

$(E.I.)_x$ emission index of specie x, g_x/kg_{fuel}

C_x concentration of specie x, ppm V

f fuel-air weight ratio, $(kg/s)_{fuel}/(kg/s)_{air}$

M_x molecular weight of specie x, $g_x/mole_x$

M_p molecular weight of combustion products, $g_{products}/mole_{products}$

The combustion efficiency was computed from the CO and HC emissions measurements. The difference between the measured and equilibrium values of these two emittants represents available chemical energy which has not been released in the combustion process. Thus, the combustion efficiency in percent is

$$EFF = 100 - 0.1\left[(E.I.)_{HC} - (E.I.)_{HC,EQ}\right] - 0.1\left[\frac{(HV)_{CO}}{(HV)_{fuel}}\right]\left[(E.I.)_{CO} - (E.I.)_{CO,EQ}\right]$$

where

EFF combustion efficiency, percent

$(HV)_x$ heating value of x, J/kg

The fuel-air ratio was determined both from the metered fuel and air flowrates and by making a carbon balance from the measured concentrations of CO, CO_2, and unburned hydrocarbons. The carbon-balance fuel-air ratio has the advantage that it is the local fuel-air ratio at which the emissions data are obtained. The equilibrium concentrations of each important specie and the adiabatic combustion temperature were obtained using the carbon-balance fuel-air ratio with the computer program of reference 15.

RESULTS AND DISCUSSION

Acceptable carbon-balance fuel-air ratios were between 90 and 112 percent of the fuel-air ratio determined from fuel and airflow measurements. Several data points (8 percent of all data taken) had values below this range and were rejected; none were above. Virtually all of those data rejected were obtained at extremely low fuel flow rates which were difficult to measure accurately. Eighty two percent of the data retained had a carbon-balance fuel-air ratio which was between 94 and 106 percent of the measured fuel-air ratio. Thus, the carbon-balance fuel-air ratio was generally a valid representation of the fuel-air ratio obtained from measured flows.

Combustion Efficiency

The combustion efficiency is shown in figure 2 as a function of the adiabatic combustion temperature. Figure 2(a) gives the results at a reference velocity of 10 m/s and for inlet temperatures of 700 to 1000 K with pressures of 3 and 6×10^5 Pa. For each set of conditions the combustion efficiency increased with adiabatic combustion temperature. At a pressure of 3×10^5 Pa the efficiency was above 99 percent for all inlet temperatures when the adiabatic combustion temperature was higher than 1200 K. At combustion temperatures below 1200 K, the effect of raising the inlet temperature was to improve the combustion efficiency.

The combustion efficiency decreased with an increase in pressure at inlet temperatures of 800 and 900 K, but it increased with pressure at 1000 K. The two different effects of pressure resulted from the relative roles of surface and gas-phase reactions. The surface reation rate is limited by that of mass diffusion, which is inversely proportional to pressure. Surface reactions cannot provide complete combustion, and gas-phase reactions, which increase with pressure, are necessary to achieve high combustion efficiency (refs. 4, 12, and 16). Thus, when the pressure is increased, combustion efficiency may either increase or decrease depending on whether gas-phase reactions become important near the reactor inlet or not until much later.

At reference velocities of 15 and 20 m/s, data were taken only with a pressure of 3×10^5 Pa (see figs. 2(b) and (c)). The effect of inlet temperature on combustion efficiency was much greater than at 10 m/s and 3×10^5 Pa. As the inlet temperature was increased, the combustion efficiency also increased.

Reference 11 reported combustion efficiencies at an inlet temperature of 800 K and a pressure of 3×10^5 Pa for the same reactor tested in this study. Higher combustion temperatures were required in the experiments of reference 11 to achieve the same efficiencies as this study. The primary reason that better performance was obtained in the present study was the elimination of most of the test section heat loss through the use of better insulation. Heat loss was calculated to be less than 2 percent, and measured exit thermocouples agreed with the adiabatic combustion temperatures within a few degrees for the present experiments. Exhaust-gas sampling techniques were different for the two experiments, as well. The single-point probe of this study was located 8 cm farther downstream than the multi-point probe of reference 11. Both of these differences would explain why higher combustion efficiencies were obtained for the present study.

Emissions

Emissions goals for the Gas Turbine Highway Vehicle Systems Project have been established (ref. 17) as 13.6 g CO/kg fuel, 1.64 g HC/kg fuel, and 1.60 g NO_x/kg fuel. These goals are used as reference values to help evaluate the measured emissions.

Carbon Monoxide

The carbon monoxide emission index is plotted as a function of the adiabatic combustion temperature in figures 3(a) to (c). Figure 3(a) gives the emissions at a reference velocity of 10 m/s for pressures of 3 and 6×10^5 Pa. Emissions decreased as the adiabatic combustion temperature increased. Values below the reference level were achieved at a pressure of 3×10^5 Pa when the adiabatic combustion temperature was higher than 1220 K for all four inlet temperatures.

As with the combustion efficiency results, the nature of the pressure effect depended on the inlet temperature. Emissions at an inlet temperature of 1000 K decreased when the pressure was doubled from 3 to 6×10^5 Pa. In contrast, at an inlet temperature of 800 or 900 K, doubling the pressure produced an increase in the CO emission index for adiabatic combustion temperatures below 1250 K, but a decrease in emissions for higher combustion temperatures.

With a reference velocity of 15 m/s (fig. 3(b)) the carbon monoxide emissions decreased with increases in either inlet temperature or adiabatic combustion temperature. The same trends are shown in figure 3(c) for a reference velocity of 20 m/s.

If figures 3(a) to (c) are compared at an inlet temperature of 1000 K it can be seen that there is little difference in the emissions produced at 10, 15, or 20 m/s reference velocities. Overall reaction rates are high enough at this inlet temperature that an increase in residence time (decrease in reference velocity) provided little benefit. Similarly, it was seen in figure 3(a) that at a reference velocity of 10 m/s there was enough residence time that an increase in inlet temperature (i.e., initial reaction rate) had little effect on the CO emissions produced.

Unburned Hydrocarbons

The emission index of unburned hydrocarbons is plotted as a function of the adiabatic combustion temperature in figure 4. Figure 4(a) gives the results at a reference velocity of 10 m/s and shows no significant effect of either inlet temperature or pressure. The temperature required to meet the hydrocarbon emission index goal was 40 K lower than that required to meet the CO emission index goal.

An effect of inlet temperature on hydrocarbon emissions was clearly seen when the reference velocity was increased to 15 m/s (fig. 4(b)). The combustion temperatures required to meet the hydrocarbon emissions goal were 30 to 80 K less than those required to meet the CO goal at this velocity.

Similarly, at 20 m/s (fig. 4(c)) emissions were higher with the lower inlet temperatures. The hydrocarbon emissions goal was achieved at combustion temperatures which were 10 to 95 K lower than those required to meet the CO goal.

As with the CO emissions, the reference velocity had little effect on the hydrocarbons emissions at an inlet temperature of 1000 K, while inlet temperature had little effect at a reference velocity of 10 m/s.

Nitrogen Oxides

The third pollutant of interest in automotive combustion studies is nitrogen oxides. Because of the low temperatures at which catalytic combustion takes place, thermal NO_x emissions are on the order of 1 ppm or less. However, conversion of nitrogen in the fuel can produce significantly higher levels than this.

The fuel used in this study contained 135 ppm of N. Eightly percent of the NO_x emissions data ranged in value from 0.2 to 0.4 g NO_2/kg fuel which represents conversion of from 45 to 90 percent of the fuel N. It should be noted, however, that there is a great deal of uncertainty associated with the measurement of such low levels of NO_x. No effect of inlet temperature, reference velocity, pressure, or adiabatic combustion temperature was apparent within the scatter of the data. The emissions were well below the reference emission index of 1.60 g NO_2/kg fuel for all test conditions.

Minimum Operating Temperature

The minimum operating temperature is the adiabatic combustion temperature above which the reactor must be operated to insure that all three pollutants meet the project goals. In this study, NO_x emissions were below the reference value at all test conditions, and the hydrocarbons goal was met at lower combustion temperatures than the CO goal. Thus, the minimum operating temperature was identical with the minimum adiabatic combustion temperature required to meet the CO goal.

Figure 5 gives the minimum operating temperature as a function of the inlet temperature. As noted in the discussion of the CO emissions, at a reference velocity of 10 m/s and a pressure of 3×10^5 Pa, excess residence time was available so that an increase in inlet temperature (and, thus, initial reaction rate) had no effect on the minimum operating temperature. A shorter reactor would probably perform equally well at these conditions.

When the pressure was increased to 6×10^5 Pa, the adiabatic combustion temperature required to meet the emissions goals increased at 800 and 900 K inlet temperatures but decreased at 1000 K. This result was due to changes in the relative roles of surface and gas-phase reactions as discussed under CO emissions.

At reference velocities of 15 and 20 m/s an increase in inlet temperature produced a decrease in the minimum operating temperature. For inlet temperatures above 1000 K the extrapolated results in figure 5 show that the minimum operating temperature approaches 1220 for all three velocities independent of inlet temperature. Thus, at an inlet temperature of 1220 K, the minimum operating temperature and the inlet temperature will be equal.

Reactor Pressure Drop

In addition to operating with very low emissions, an automotive gas turbine combustor must have minimal pressure losses to permit high cycle efficiencies. In general, the pressure drop for a catalytic reactor includes both friction losses and entry/exit losses. For the reactor and the test conditions of this study, however, friction losses dominated. The Reynolds number was less than 1000 for all flow conditions; thus, the flow through the reactor passages was laminar. For laminar flow, the friction drop over a small element of length, $d\ell$, is

$$\frac{dp}{p} = k \frac{VT^{0.6}}{p} d\ell$$

where

k a constant

V local velocity

T local temperature

p pressure

The local velocity is equal to the reference velocity V_{ref}, times the ratio of the local to inlet temperature, T/T_{in}. Then the pressure drop over the reactor length, ℓ, is

$$\frac{\Delta p}{p} = \frac{k}{T_{in}} \frac{V_{ref}}{p} \int_0^{\ell} T^{1.6} d\ell$$

The pressure drop as a percent of upstream pressure is presented in figure 6 as a function of V_{ref}/p. For an inlet temperature of 700 K the data show a direct proportionality between $\Delta p/p$ and V_{ref}/p. This result suggests that the temperature history, $T = f(\ell)$, does not change appreciably with reference velocity. At higher inlet temperatures, some decrease in pressure drop was observed as would be expected for laminar flow friction loss. For example, at a reference velocity of 10 m/s and a pressure of 3×10^5 Pa

$\left(V_{ref}/p = 3.3\times10^{-5}\ \frac{m/s}{Pa}\right)$, the pressure drop decreased from 0.8 to 0.66 percent when the inlet temperature was raised from 700 to 1000 K.

The pressure drop data in figure 6 at an inlet temperature of 800 K and a pressure of 3×10^{5} Pa were about half the values reported in references 11 and 14 for the same reactor. In those previous studies, however, the thermocouple instrumentation within the reactor housing was more dense than that used for the present study. In addition, the earlier data included the pressure drop across the downstream instrumentation section. For the very low pressure drops at which this reactor operates, these additional instrumentation pressure drops were significant.

CONCLUSIONS

This study of the effect of inlet temperature on the performance of a catalytic reactor has shown that if sufficient residence time was provided, as occurred at a reference velocity of 10 m/s, inlet temperatures in the range 700 to 1000 K had little effect on combustion efficiency or emissions. At 700 K inlet temperature, significantly better performance was achieved at 10 m/s than at higher reference velocities. Thus, for applications with low inlet temperatures, such as the aircraft or stationary gas turbines, long residence times (low reference velocities) must be provided to achieve high combustion efficiencies.

In contrast, the reference velocity had little effect on the combustion efficiency or emissions when the inlet temperature was 1000 K. This result suggests that for combustor applications with high inlet temperature, such as the regenerative automotive gas turbine, the upper limit on reference velocity may be determined by permissible pressure drop rather than combustion efficiency.

The pressure drop in this study resulted primarily from friction loss through the reactor passages. Because the flow was laminar, the percent pressure drop decreased slightly with increasing inlet temperature at a constant adiabatic combustion temperature.

The improved catalyst performance which results from operation at high in-inlet temperatures makes the automotive gas turbine an especially attractive application for catalytic combustion. In this study, at 1000 K inlet temperature, 1300 K exit temperature, and 20 m/s reference velocity, emissions of CO, unburned hydrocarbons, and NO_x were below levels which would permit the most stringent automotive standards to be achieved, and pressure drop was only 1.5 percent.

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TABLE I. - DESCRIPTION OF CATALYST ELEMENTS

Element number	1	2
Element designation	JM1	JM2
Position in reactor	Upstream	Downstream
Manufacturer	Johnson Matthey, Ltd.	Johnson Matthey, Ltd.
Catalyst	Pt	Pd
Loading, kg/m^3	5.3	5.3
Substrate	Metal foil, corrugated and wound into a cylinder	Metal foil, corrugated and wound into a cylinder
Cell density, $cells/cm^2$	62	62
Element diameter, cm	12	12
Element length, cm	7.6	7.6

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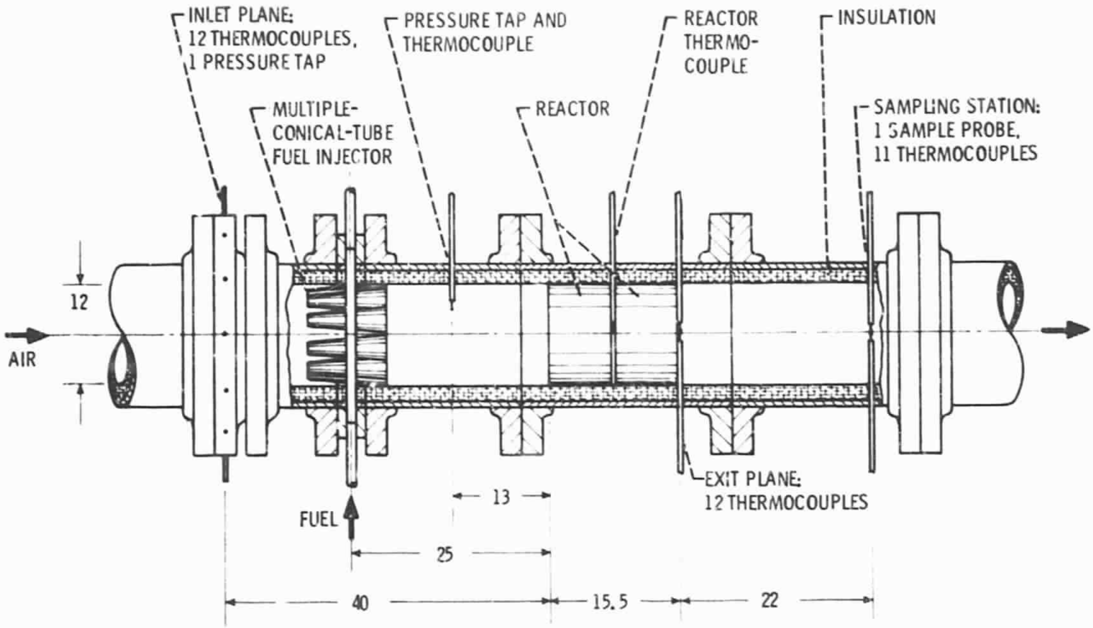
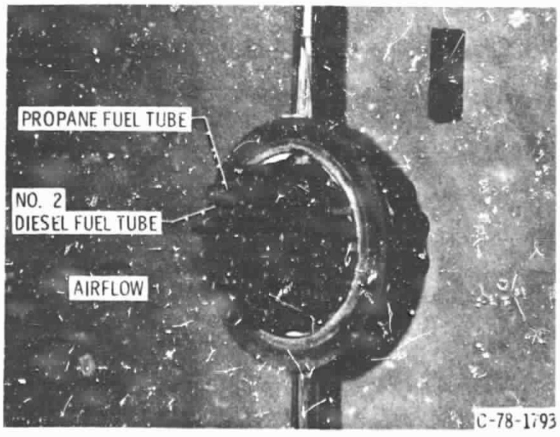


(a) TEST SECTION (DIMENSIONS IN cm).

Figure 1. - Experimental apparatus.



(b) MULTIPLE CONICAL TUBE FUEL INJECTOR.

Figure 1. - Concluded.

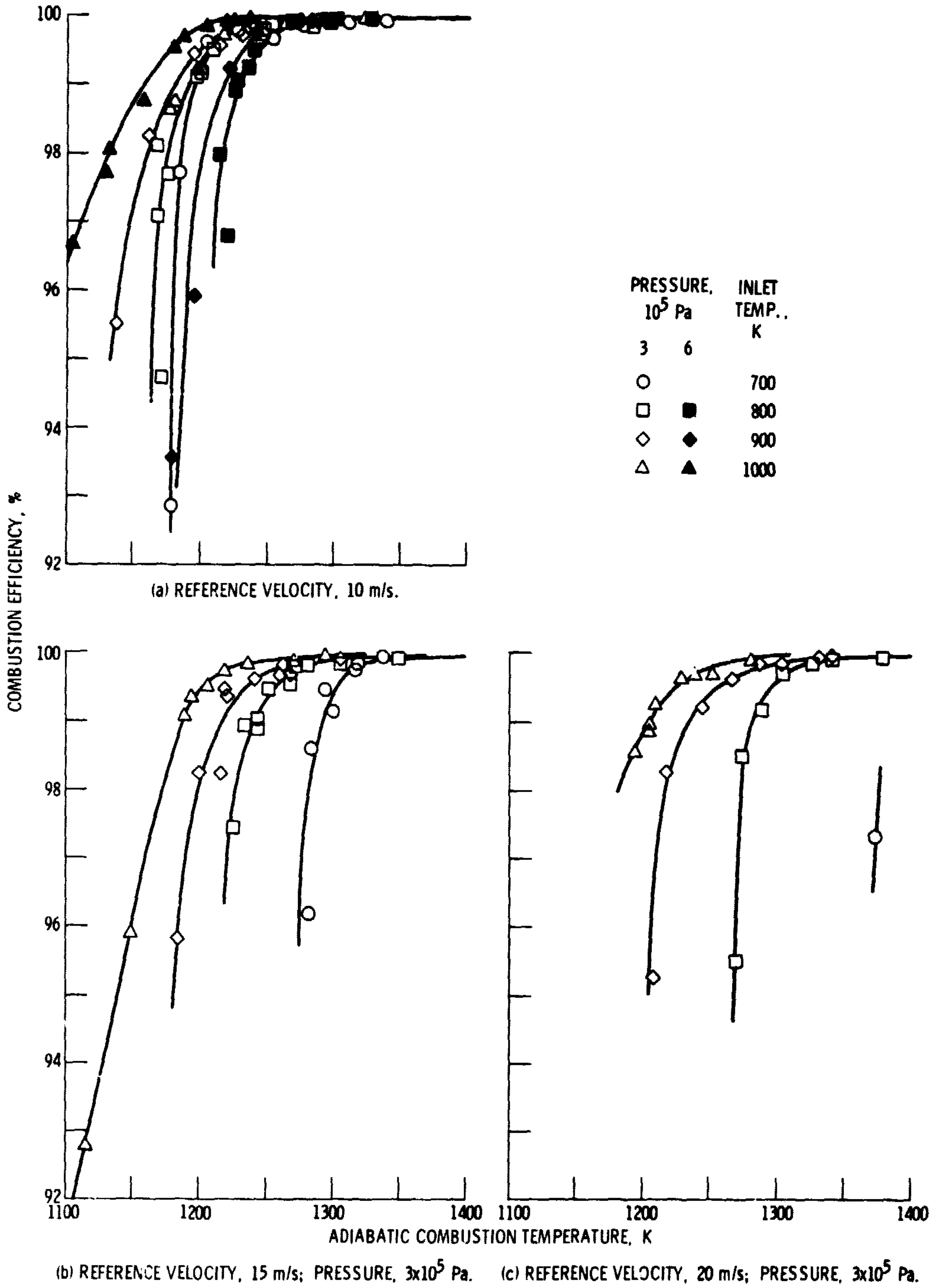


Figure 2. - Combustion efficiency.

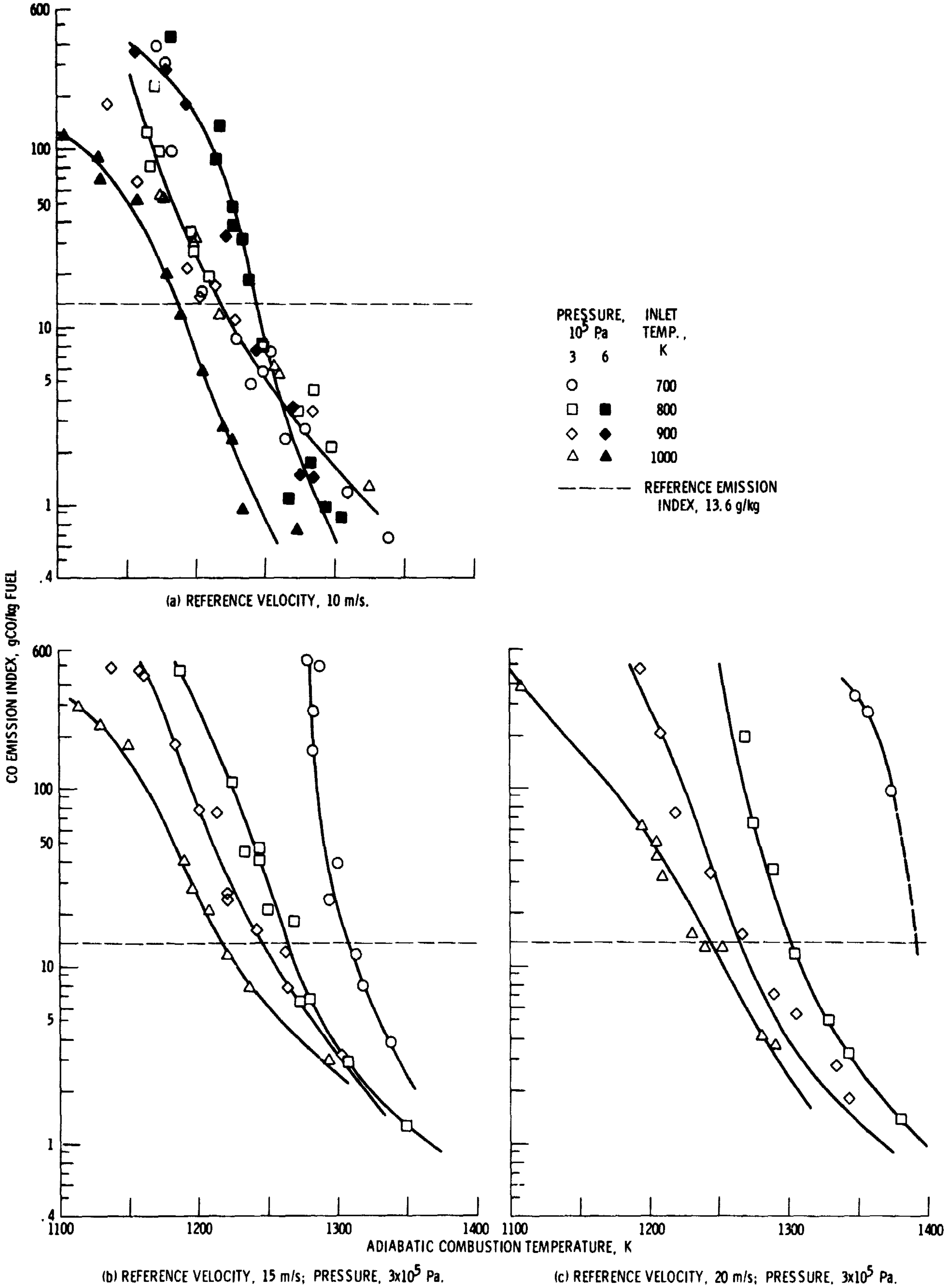


Figure 3. - Carbon monoxide emissions.

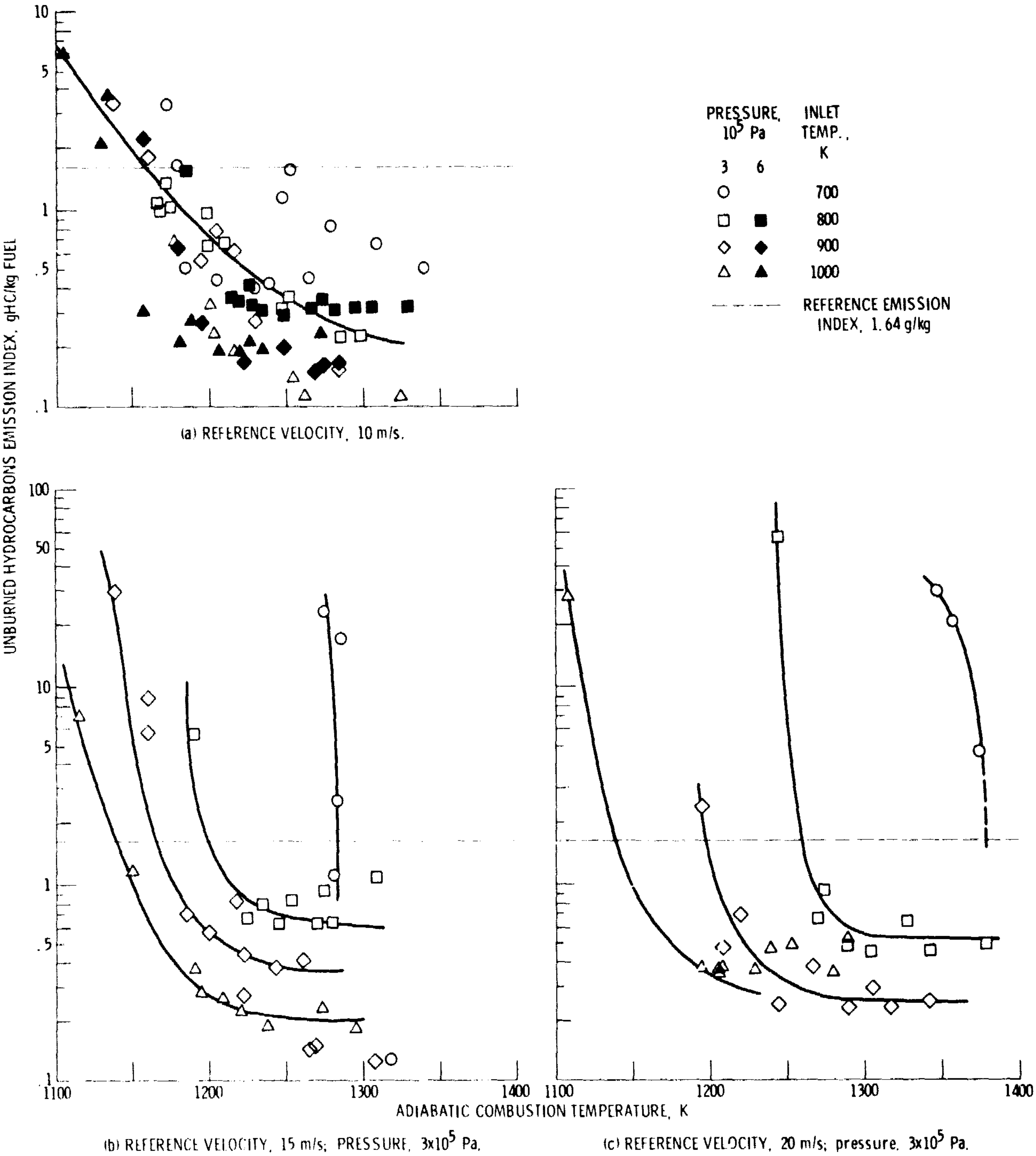


Figure 4. - Unburned hydrocarbons emissions.

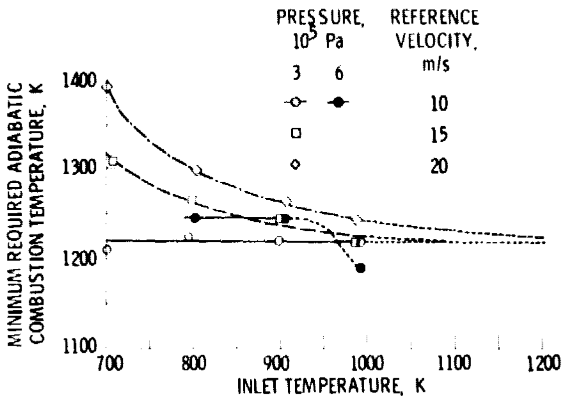


Figure 5. - Effect of inlet temperature on minimum operating temperature required for low emissions.

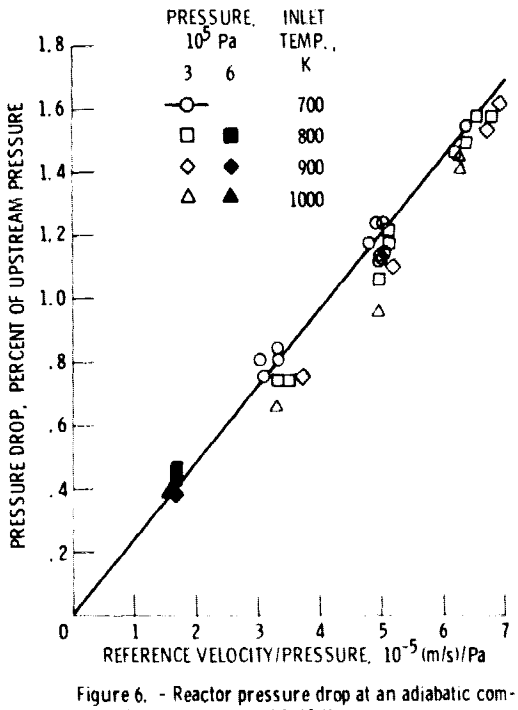


Figure 6. - Reactor pressure drop at an adiabatic combustion temperature of 1300 K.