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EMISSIONS OF OXIDES OF NITROGEN FROM AN EXPERIMENTAL PREMIXED-HYDROGEN BURNER

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16. Abstract

Flame-tube experiments using premixed hydrogen and air were conducted to determine the emissions of oxides of nitrogen (NO_x) resulting from ultralean combustion. Measurements of NO_x emissions and combustion efficiency were made for inlet mixture temperatures of 600 and 700 K, pressures of 3.8×10^5 and 5.2×10^5 N/m^2, reference velocities of 15 to 18 m/sec, and equivalence ratios of 0.2 to 0.4. At the 700 K inlet mixture temperature, NO_x emissions were 0.06 ppmv, and combustion efficiency was 98 percent at an equivalence ratio of 0.24. The use of a high-blockage (92-percent blockage) flameholder made it possible to conduct tests without upstream burning in the premixing duct for mixtures with equivalence ratios less than 0.4. For richer mixtures upstream burning did occur and prevented further testing.

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EMISSIONS OF OXIDES OF NITROGEN FROM AN EXPERIMENTAL PREMIXED-HYDROGEN BURNER

by David N. Anderson

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SUMMARY

Emissions of oxides of nitrogen (NO_x) were measured in an experimental flame tube burning premixed hydrogen and air to determine if the use of hydrogen fuel would result in reduced NO_x emissions compared with hydrocarbon fuels. Inlet mixture temperatures of 600 and 700 K, burner pressures of 3.8×10^5 to 5.2×10^5 newtons per square meter, and reference velocities of 15 to 18 meters per second were tested. Emissions were measured over a range of hydrogen equivalence ratios of 0.2 to 0.4 with corresponding flame temperatures of 1250 to 1700 K.

As expected, NO_x concentration was strongly dependent on flame temperature. As a result, hydrogen has the potential for low NO_x emissions because it can be burned at lower temperatures than hydrocarbon fuels. The lean blowout limit for hydrogen in these tests was 1150 K with a 600 K inlet mixture temperature. The corresponding equivalence ratio was 0.18.

With a 700 K inlet mixture temperature the minimum equivalence ratio which gave a combustion efficiency of 98 percent or greater was 0.24 (flame temperature of 1380 K). At this condition, the NO_x concentration was 0.06 part per million by volume (ppmv). At the 600 K inlet mixture temperature NO_x concentrations were as high as 1.2 ppmv at an equivalence ratio of 0.39 (flame temperature of 1650 K). For the conditions tested (equivalence ratio less than 0.4), NO_x emissions were independent of residence time over the range 0.4 to 2.9 milliseconds.

The maximum equivalence ratio tested was limited by flashback of the flame into the premixing duct. This problem of flashback may present the greatest obstacle to the development of a practical premixed-hydrogen combustor.

INTRODUCTION

In an experimental flame tube burning premixed hydrogen and air, concentrations of oxides of nitrogen (NO_x) were measured to determine what emission benefits would result if hydrogen were used as a fuel in a gas turbine combustor.

Except for NO_x formation from fuel-bound nitrogen, the rate of NO_x formation is exponentially dependent on local flame temperature; thus, reducing the flame temperature is an effective way to lower NO_x emissions. This reduction can best be achieved both by eliminating local hot spots and by lowering the average flame temperature. Several studies using different hydrocarbon fuels (refs. 1 to 4) have shown that NO_x emissions can be reduced by about an order of magnitude compared with those from conventional gas turbine combustors when lean, premixed, prevaporized fuel-air mixtures are burned. The minimum NO_x levels which can be achieved by lean burning are limited by the lean flammability limit of the fuel, which occurs at a flame temperature of about 1800 K for most hydrocarbon fuels.

The lean flammability limit for hydrogen is reported in reference 5 to occur at a flame temperature of 1060 K with a 300 K inlet mixture temperature and at 1140 K with a 600 K inlet mixture temperature. Thus, the addition of small quantities of hydrogen to a hydrocarbon should permit leaner burning and therefore lower NO_x formation than is possible with the pure hydrocarbon. In test programs reported in references 6 and 7 hydrocarbon flames were enriched with small quantities of hydrogen. In the study of reference 6 up to 4 percent hydrogen by weight was injected into an experimental combustor designed for operation with liquid Jet A fuel. Because the combustor design did not provide a uniform fuel-air mixture, little effect of hydrogen injection on emissions was observed. In the study of reference 7 up to 5 percent hydrogen was injected into a premixed prevaporized propane flame. The minimum flame temperature for which combustion efficiency was at least 99 percent decreased from 1880 K with no hydrogen to 1760 K with 4 percent hydrogen. The corresponding NO_x emissions were lowered by nearly a factor of 2 at an inlet temperature of 700 K.

More dramatic reductions in NO_x should be possible if stable burning can be achieved at still lower flame temperatures. Hydrogen has the low flame temperature capability and also has the advantage over hydrocarbon fuels that combustion produces no carbon monoxide, unburned hydrocarbons, smoke, or sulfur dioxide. An investigation of the potential of hydrogen for reduced emissions is reported in reference 8. Minimum primary zone equivalence ratio tested was computed to be 0.43, but because the combustor did not burn a homogeneous fuel-air mixture, only a 22-percent reduction in NO_x emissions was observed compared with values from a reference hydrocarbon-fueled combustor with a computed primary zone equivalence ratio of 0.62.

In the study described in this report the flame-tube apparatus of reference 7 was used. Hydrogen was premixed with air upstream of the burner to achieve a homogeneous fuel-air mixture. A perforated plate with 92 percent blockage was used to stabilize the flame. Test conditions included inlet mixture temperatures of 600 and 700 K, burner pressures of 3.8×10^5 to 5.2×10^5 newtons per square meter, and reference (burner inlet) velocities of 15 to 18 meters per second. Measurements were made of exit temperature and total NO_x concentrations.

APPARATUS AND PROCEDURE

Figure 1 is a schematic of the test hardware. Hydrogen was injected into an indirectly preheated airstream 2.6 meters upstream of the flameholder.

To avoid a flame-holding surface in the inlet duct a single Chromel-Alumel thermocouple was used instead of a thermocouple rake to measure the inlet mixture temperature. It was placed about 0.10 meter upstream of the flameholder and extended into the duct about 1 centimeter from the duct wall. Because the hydrogen entered the airstream at about 300 K and had to be heated to 600 or 700 K by the air, the inlet mixture temperature measured by this thermocouple was computed to be from 30 to 53 K less than the inlet air temperature, depending on the hydrogen flow rate, for these experiments.

The water-cooled flameholder (fig. 2) was made from two stainless-steel plates with eighty 0.318-centimeter-inside-diameter tubes providing the passage. The flameholder was water cooled to safeguard against burnout, which had been experienced in previous tests using perforated-plate flameholders (ref. 1). This configuration had a blockage of 92 percent of the duct cross-sectional area. This high blockage was necessary to prevent propagation of the hydrogen flame front upstream of the flameholder. The effect of flameholder design on performance was not made a part of this study; however, combustion stability was enhanced and the tendency of the flame to flash upstream into the premixing duct was lessened by allowing the tubes in the flameholder to project about 1 centimeter from the downstream face of the flameholder and flaring them.

The water-cooled burner (see fig. 1) was 0.103 meter in diameter, as was the inlet duct, and 0.31 meter long. The pressure drop across the burner was measured with single wall static taps, one located upstream of the flameholder and one downstream of the exit thermocouple rake. Because the dynamic pressure amounted to only about 0.2 percent of the total pressure for the conditions of this experiment, the measured static-pressure drop could be considered to be equivalent to the total-pressure drop. An array of eight platinum/platinum-13-percent-rhodium thermocouples, arranged as shown in figure 1, was used to measure exit temperature.

Combustion products on the burner centerline were sampled with a single-point water-cooled gas sampling probe. Details of the probe design are shown in figure 1. The probe could be traversed axially along the burner centerline to vary the combustion residence time at which sampling occurred. The use of a centerline probe should have ensured that the sample was free from wall-cooling effects and that the NO_x concentrations measured were representative of adiabatic combustor conditions.

Unheated nylon tubing with a dry-ice water trap connected the sample probe to the chemiluminescent NO_x analyzer. Both total NO_x (NO plus NO_2) and NO could be measured with this instrument; however, only the total NO_x values are reported.

Hydrogen flow rate was measured by using a calibrated venturi. Some properties of hydrogen are given in table I. Airflow was measured by using an ASME standard orifice.

RESULTS AND DISCUSSION

Emissions of Oxides of Nitrogen

Emissions of NO_x were measured for both 600 and 700 K inlet mixture temperatures and 3.8×10^5- and 5.2×10^5-newton-per-square-meter burner pressures. The bulk of the data was taken with a constant combustion residence time of about 1 millisecond before sampling. The residence time was computed by using the probe position, the adiabatic flame temperature, and the burner pressure. The probe position could be varied, as combustor conditions changed, to maintain a constant time. A number of test points were also taken at several different conditions to determine the effect of residence time on NO_x emissions. In spite of time variations of more than a factor of 5, no effect of residence time could be seen in the NO_x emissions within the experimental scatter. This result confirms the observation of reference 1 that, because of superequilibrium oxygen atom concentration at lean conditions, as equivalence ratio is reduced, residence time has less and less effect on NO_x levels. Because residence time showed no measurable influence, the results are given for all residence times tested.

All measurements were made by using a full-scale analyzer range of 2.5 parts per million by volume (ppmv), the lowest range available. The instrument should be accurate to 2 percent of the full-scale value, or 0.05 ppmv. Background levels of NO_x were measured before each test run. They varied between 0.05 and 0.15 ppmv and were subtracted from the test readings to obtain reported values of combustion-generated NO_x. Measured levels which exceeded the background concentration by less than 0.05 ppmv are not reported. Such values were observed when the equivalence ratio was less than 0.24.

The NO_x emissions are presented in figure 3. Figure 3(a) reports the emission index (g NO_2/kg fuel burned) as a function of the hydrogen equivalence ratio. The emission index is given in two forms. In the first, it is based on the weight of propane required to give the same enthalpy rise as the hydrogen actually burned, that is, the equivalent weight of propane. This form permits direct comparison of the results with earlier test results which used propane fuel (refs. 1 and 7). Because the heating value of propane is close to that of other hydrocarbon fuels, the equivalent-propane emission index also allows close comparison with results obtained by using other hydrocarbon fuels. The second form of emission index given is based on the actual weight of hydrogen burned. Emission data obtained for conditions yielding less than 98 percent combustion efficiency are shown as tailed symbols in figure 3 to indicate undesirable operating conditions.

The results show the strong influence of equivalence ratio on emissions. At the 600 K inlet mixture temperature, NO_x emissions ranged from about 0.007 gram of NO_2 per kilogram of equivalent propane at an equivalence ratio of 0.3 to about 0.07 gram of NO_2 per kilogram of equivalent propane at an equivalence ratio of 0.4. For the 700 K inlet temperature the range of equivalence ratio from 0.23 to 0.26 resulted in NO_2 emission indices between 0.004 and 0.006 gram of NO_2 per kilogram of equivalent propane.

In agreement with the trend shown in figure 3(a), NO_x emissions from experimental premixed-hydrocarbon-fueled burners (refs. 1 and 4) also decreased exponentially as equivalence ratio was reduced. However, because of lean blowout, conditions could not be made lean enough in those studies to obtain NO_x emissions below 0.2 to 0.3 gram of NO_2 per kilogram of fuel.

The effect of flame temperature on NO_x volumetric concentration is shown in figure 3(b). The curve through the data is the prediction of a well-stirred-reactor computer model (ref. 9) using a mixture of propane and hydrogen as fuel.

The measured NO_x concentration ranged from values which were too low to be measured accurately (less than 0.05 ppmv) at flame temperatures below 1300 K to levels as high as 1.2 ppmv at flame temperatures of nearly 1700 K. Fairly good agreement between the well-stirred-reactor predictions and the experimental results is indicated by figure 3(b) for temperatures above 1550 K. For lower temperatures, where the data scatter and uncertainty are greatest, the measured values are higher than the predicted results. The bulk of the data does agree with predicted orders of magnitude, however.

The well-stirred-reactor reference curve is a lower temperature portion of the analytical curve shown in figure 4 of reference 7. In the study of reference 7 the experimental data agreed with both the trend and magnitudes of the NO_x concentration predicted by this analytical model when either propane fuel or various mixtures of propane and hydrogen were burned. In the present study, when hydrogen fuel was used, NO_x concentrations again agreed fairly well, for a lower flame temperature region, with the same predictions. This result supports the conclusion of reference 7 that NO_x concentration is independent of the fuel burned and, over the range of inlet temperatures from 600 to 700 K, depends only on flame temperature when the pressure and residence time are held constant. Thus, if it were possible to burn nitrogen-free hydrocarbon fuels at the lean conditions considered in this study, NO_x concentrations would be the same as those measured from hydrogen combustion. The NO_x concentrations recorded were significantly lower than those obtained in premixing studies using hydrocarbon fuels only because hydrogen can be burned with much lower flame temperatures than can hydrocarbon fuels.

No effect of pressure was seen in this study for the pressure range from 3.9×10^5 to 5.2×10^5 newtons per square meter.

Pressure Drop and Combustion Stability

The flame speed for hydrogen is nearly seven times that of hydrocarbon fuels (see table I); therefore, it is necessary to maintain high velocities through the flameholder to prevent the flame from propagating upstream into the premixing duct. Unfortunately, high velocities result in large pressure drops. As mentioned in the section APPARATUS AND PROCEDURE, a flameholder was used in this study which had a blockage of 92 percent in order to prevent propagation of the flame upstream through the flameholder. The pressure drops which resulted at each of the test conditions ranged from 25 to 41 percent of the upstream pressure (as shown in table II). Although these values are much higher than those that would be acceptable in a practical combustor, it is not expected that combustor pressure drop would have a significant effect on NO_x emissions.

Even with the high-blockage flameholder, flashback of the flame occurred when the equivalence ratio was increased above 0.43 with the 600 K inlet temperature or 0.26 with the 700 K inlet temperature. These conditions corresponded to maximum flame temperatures of 1700 K with the 600 K inlet and 1450 K with the 700 K inlet.

Lean blowout of the flame resulted for equivalence ratios below 0.18 when the inlet mixture temperature was 600 K. The corresponding flame temperature was 1150 K. The lean blowout limit for 700 K was not established because the fuel flow rate at blowout was too low to be measured accurately with the metering installation used. Data were recorded at flame temperatures as low as 1260 K (equivalence ratio of 0.18), however.

Combustion Efficiency

The combustion efficiency was computed from the exit thermocouple measurements as explained in the appendix. The results are shown in figure 4. Because the thermocouple rake was fixed in position, it was not possible either to vary the combustion residence time for a given test condition or to maintain a constant residence time for all conditions. The variation in test conditions resulted in the ranges of residence time shown in figure 4. This time is based on the airflow rate, the burner pressure, the adiabatic flame temperature, and the distance from the flameholder to the thermocouple rake (0.31 m). It is not expected that the time variations shown would have a significant effect on combustion efficiency.

Combustion efficiency increased rapidly with equivalence ratio and approached 100 percent for equivalence ratios greater than 0.24 with the 700 K inlet mixture temperature and 0.31 with the 600 K inlet mixture temperature. Because the combustion efficiency was determined from the exit thermocouple measurements, the actual values depended on assumptions used in making radiation corrections to the thermocouple readings (see the appendix). Depending on the assumed effective wire diameter and wire emissivity, the combustion efficiency could vary by 3 to 4 percent. Combustion efficiencies near 100 percent could be expected at the higher equivalence ratios, however, so the computed values appear to be reasonable. The small change in burner pressure from 3.8×10^5 to 5.2×10^5 newtons per square meter had no apparent effect on combustion efficiency.

CONCLUDING REMARKS

The tests described in this report demonstrated the ultralow NO_x emissions that are possible from a hydrogen-fueled experimental combustor. Combined with the lack of emissions of either carbon monoxide or unburned hydrocarbons, this low NO_x production makes hydrogen potentially a very attractive fuel from an ambient air quality viewpoint.

In order to realize the ultralow NO_x potential of hydrogen, however, it is necessary to burn with a very lean, homogeneous mixture of hydrogen and air. A previous study (ref. 8) shows that without premixing to achieve a homogeneous mixture, NO_x emissions from a hydrogen-fueled combustor were comparable with those from a hydrocarbon-fueled combustor. The advantage of hydrogen results only from the capability to burn with lower flame temperatures than are possible with hydrocarbon fuels; elimination of hot regions which result from nonhomogeneous mixtures is essential.

This test program also emphasized one of the main problems that faces designers of a practical premixed-hydrogen-fueled combustor. In these tests, flashback of the flame into the inlet duct was a constant problem. It was alleviated only by resorting to a flameholder with impractically high blockage. Even with this flameholder, flashback occurred when the mixture was richer than a 0.4 equivalence ratio. A possible solution is the use of the swirl-can combustor described in reference 10 because it does not require a mixing duct upstream of the flameholder. Whether or not it can provide the uniformity of mixture required for low NO_x production remains to be shown, however.

These experiments have demonstrated both the potential of hydrogen fuel for ultraclean combustion and the difficulty of designing a practical combustor which could utilize hydrogen fuel.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, March 11, 1976,
505-03.

APPENDIX - COMPUTATIONAL METHODS

Emission Index

The emission index (g pollutant/kg fuel burned) is a measure of the quantity of pollutant formed relative to the amount of fuel burned. It is determined from the measured pollutant concentration by using the formula

$$EI_x = \frac{M_x}{M} \frac{1+f}{f} C_x \times 10^{-3}$$

where

EI_x	emission index of pollutant x, g pollutant/kg fuel
M_x	molecular weight of pollutant x
M	molecular weight of combustion products
f	fuel-air weight ratio
C_x	concentration of pollutant x, ppmv

In this study, the only pollutant measured was NO_x. It is customary to report total NO_x in terms of grams of NO_2 per kilogram of fuel: thus, M_x is the molecular weight of NO_2, 46.

The molecular weight of combustion products was determined from the following expression:

$$M = 28.97 \frac{1+f}{1+7.24 f}$$

where 28.97 is the molecular weight of air, the factor 7.24 is one-half the ratio of the molecular weight of air to that of hydrogen, and f is the hydrogen-air weight ratio.

The value of C_x is the actual concentration of pollutant in ppmv. Because the sample was dried before it was analyzed for NO_x in this study, the measured concentration had to be corrected for the water vapor removed in order to obtain the true, or wet-basis, concentration C_x. The water vapor removed from the sample line was assumed to be that which resulted from complete combustion of hydrogen in air. The actual (wet) concentration is related to the measured (dry) concentration C_{DRY} as follows:

$$C_x = C_{DRY} \frac{1 - 7.24 f}{1 + 7.24 f}$$

Again, the factor 7.24 is one-half the ratio of the molecular weight of air to that of hydrogen, anf f is the hydrogen-air weight ratio.

For comparison with results from combustion with hydrocarbon fuels, it is convenient to express the emission index in terms of an equivalent weight of hydrocarbon burned (g pollutant/kg equivalent hydrocarbon). The hydrocarbon-equivalent emission index is the emission index which would result if the same pollutant concentration were formed while burning a hydrocarbon fuel at the flow rate necessary to give the same enthalpy rise as hydrogen. The ratio of lower heating value of hydrogen to that of propane is 2.59; therefore, the propane-equivalent emission index is $EI_{\text{equivalent propane}} = EI_x/2.59$.

Radiation Correction

Because of the presence of cooled walls, the exit thermocouples were corrected for radiation loss according to the expression (ref. 11)

$$T_{4,C} = T_{4,\text{meas}} + \frac{K}{\sqrt{Mp}} \left(\frac{T_{4,\text{meas}}}{556}\right)^{-0.18} \left[\left(\frac{T_{4,\text{meas}}}{556}\right)^4 - \left(\frac{T_w}{556}\right)^4\right]$$

where

$T_{4,C}$	actual (corrected) gas temperature, K
$T_{4,\text{meas}}$	thermocouple reading, K
K	radiation correction coefficient, K
M	Mach number at thermocouple
p	pressure at thermocouple, atm
T_w	wall temperature, K

The wall temperature was nearly constant at 300 K for these experiments.

The radiation correction coefficient K for a bare wire in crossflow has the form (ref. 11)

$$K = 15\,\epsilon_w \sqrt{\frac{D}{2.54}}$$

where

ϵ_w emissivity

D effective wire diameter, cm

For clean platinum/platinum-13-percent-rhodium wire the emissivity has been experimentally determined (ref 12) to depend on wire temperature:

$$\epsilon_w = 0.09 + 0.04\,\frac{T_{4,meas}}{556}$$

However, for dirty wires, the emissivity can be as much as three times this value (ref. 12). Therefore, for correcting the temperature measurements of this study, an emissivity of

$$\epsilon_w = 3\left(0.09 + 0.04\,\frac{T_{4,meas}}{556}\right)$$

was used.

Thermocouples having 0 051-centimeter-diameter wires were used in this study. An effective diameter D of 0.071 centimeter was used to allow for the weld bead at the thermocouple junction.

Combustion Efficiency

The combustion efficiency is given by the following expression:

$$\eta = \frac{T_{4,c} - T_3}{T_{4,eq} - T_3}$$

where

$T_{4,C}$	average exit temperature
T_3	inlet mixture temperature
$T_{4,eq}$	equilibrium exit temperature

The average exit temperature was obtained from the eight thermocouple readings by applying the radiation correction to the average value. A more accurate approach would be to correct each measured temperature before averaging; however, the difference between the maximum and the minimum temperature of the exit thermocouple array was never more than 7.5 percent, and for the most readings there was less than 5 percent variation.

The equilibrium exit temperature was computed from the adiabatic flame temperature (determined by using the computer program of ref. 13) by subtracting the loss due to water cooling of the burner. This heat loss was found by using cooling flow measurements and the measured water temperature increase across the burner. It varied between 7.5 and 10.5 percent of the ideal enthalpy rise from combustion.

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TABLE I. - PROPERTIES OF HYDROGEN FUEL

Analysis	
Hydrogen, percent	99.94
Oxygen, ppmv	5
Carbon monoxide, ppmv	5
Carbon dioxide, ppmv	5
Methane, ppmv	5
Water vapor, ppmv	1.5
Organic chlorides, ppmv	1.0
Nitrogen	Bal.
Lower heating value, J/g	120 000
Lower heating value relative to propane	2.59
Flammability limits at 673 K (ref. 5)	
Lean equivalence ratio	0.16
Rich equivalence ratio	10.5
Stoichiometric fuel-air ratio	0.029
Flame speed relative to propane	6.8

TABLE II. - PRESSURE DROP

Inlet mixture temperature, K	Burner pressure, N/m^2	Reference velocity, m/sec	Pressure drop, percent of upstream pressure
600	3.8×10^5	17	41
600	5.2	15	25
700	3.8	18	35

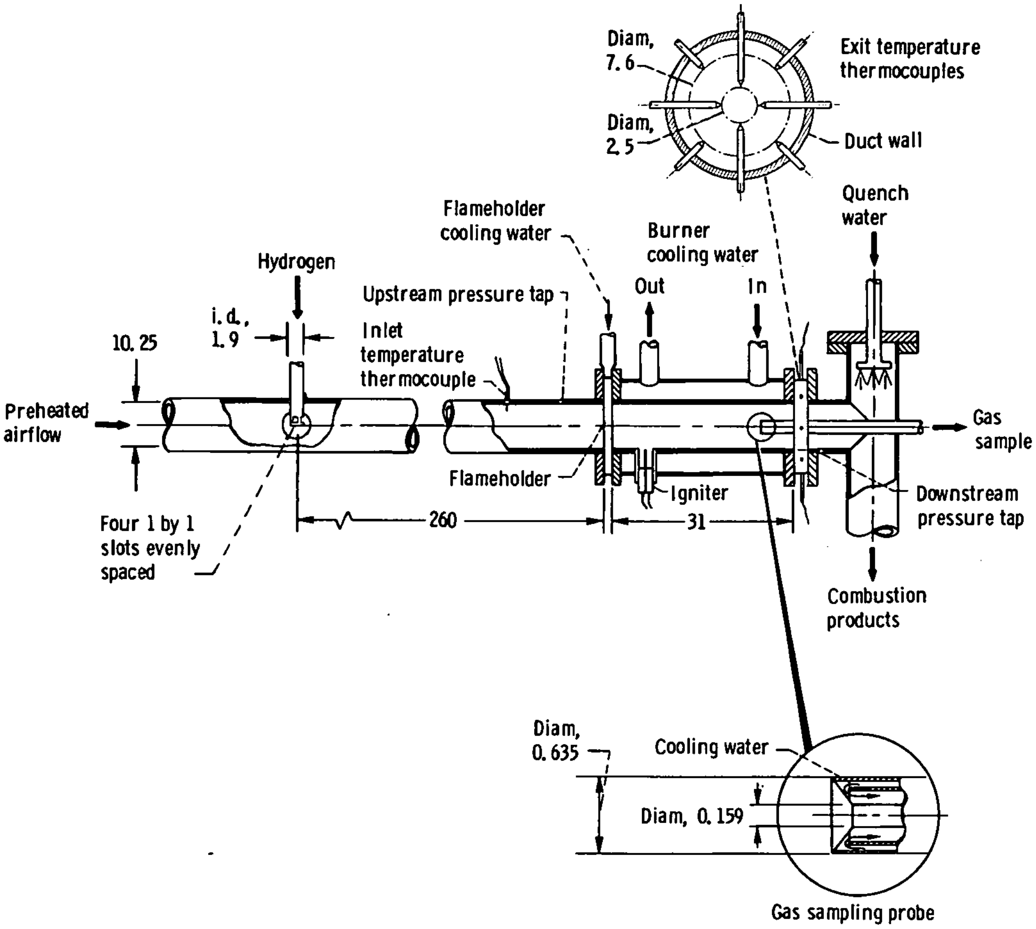

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Figure 1. - Rig schematic. (Dimensions are in centimeters.)

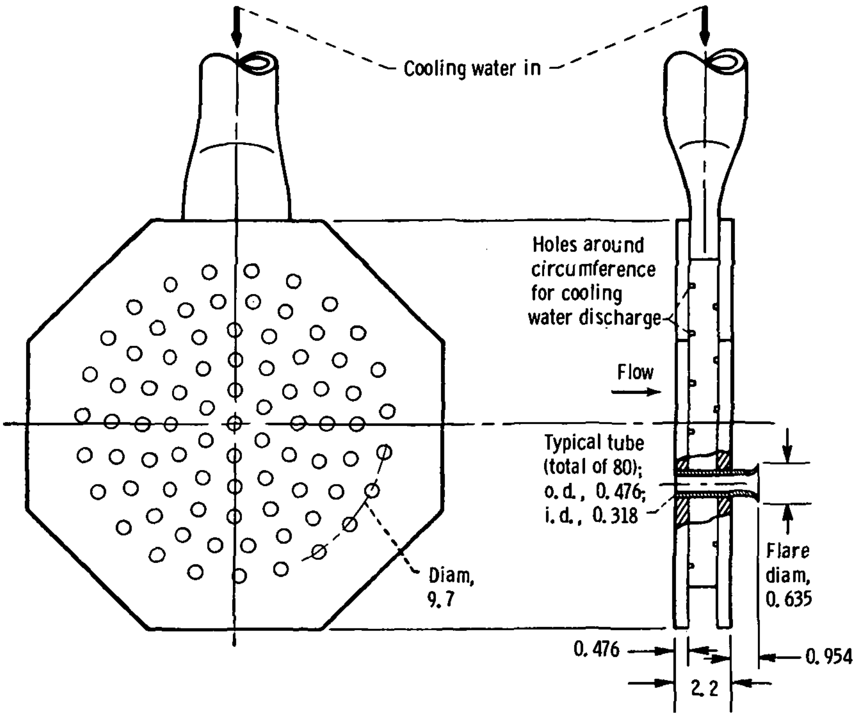
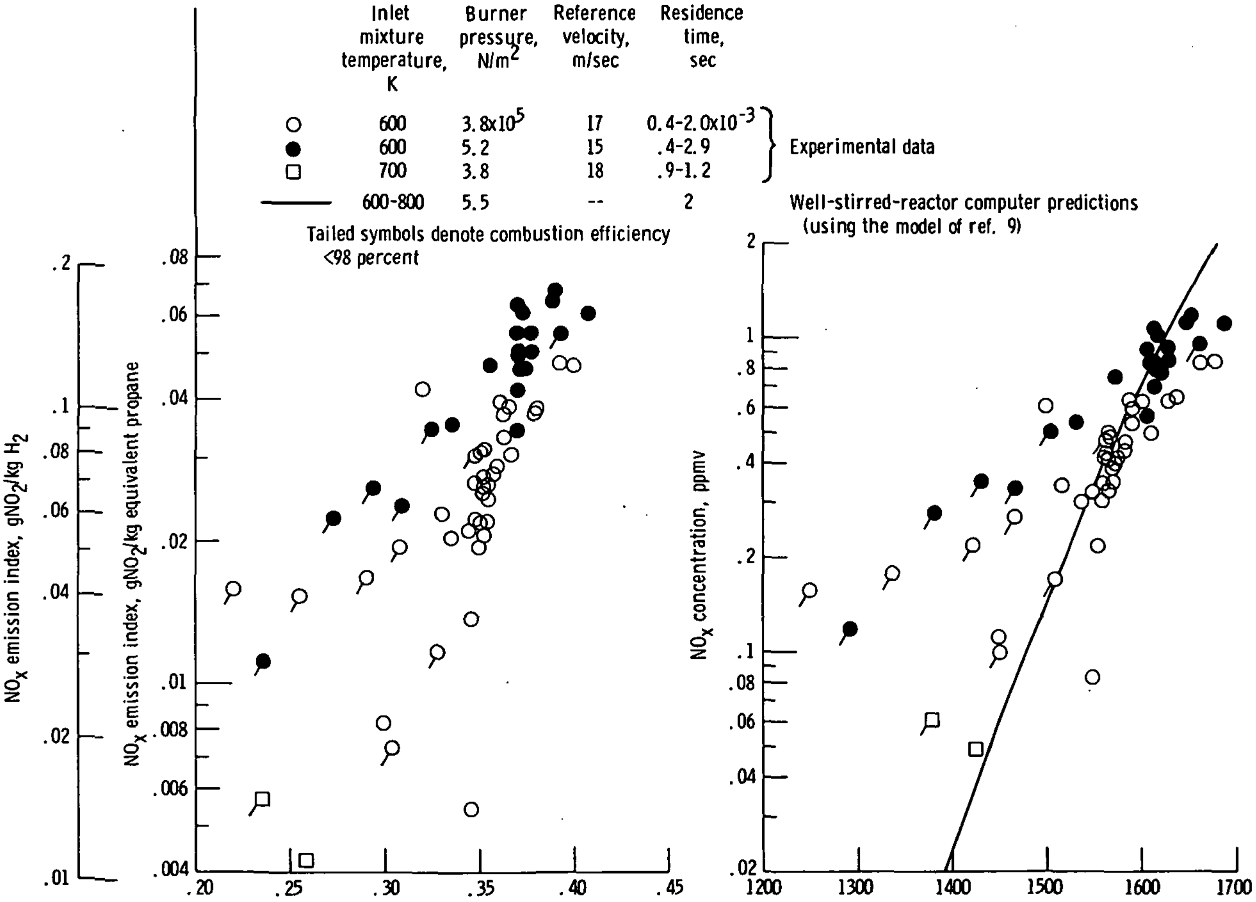


Figure 2. - Flameholder. Open area, 8 percent. (Dimensions are in centimeters.)



(a) Emission index.

(b) Effect of flame temperature on total concentration of oxides of nitrogen.

Figure 3. - Emissions of oxides of nitrogen.

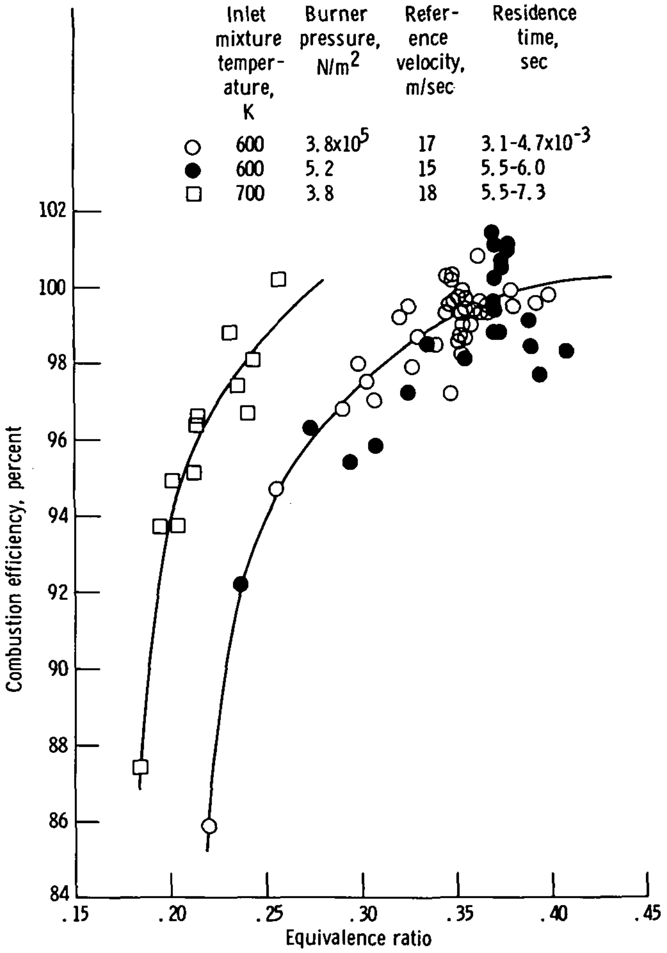


Figure 4. - Combustion efficiency.

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