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Catalytic Combustion With Steam Injection

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David N. Anderson and Robert R. Tacina
Lewis Research Center
Cleveland, Ohio



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CATALYTIC COMBUSTION WITH STEAM INJECTION

by David N. Anderson and Robert R. Tacina

National Aeronautics and Space Administration
Lewis Research Center
Cleveland, Ohio

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INTRODUCTION

This report describes an experimental study to determine: (1) if catalytic combustion performance is degraded when steam is injected into the airstream, and (2) if steam-assisted fuel injection might eliminate the upstream burning problems which have occurred in past studies of catalytic combustion of residual fuels.

The steam-injected cycle has been proposed for stationary gas turbines as a way to reduce fuel consumption by recovering waste exhaust heat (1 to 5). Figure 1 describes this cycle, and shows how the steam is generated from waste heat in the turbine exhaust and then is injected into the combustor. Figure 2, taken from the calculated results of (1), illustrates how steam injection improves the cycle efficiency for a nonregenerative cycle. The cycle efficiency is plotted as a function of the turbine-inlet temperature for steam injection rates as high as 23 percent of the compressor discharge air flow rate and for compressor pressure ratios of 8 and 16. The steam temperatures and pressures used in the analysis of (1) were based on what could realistically be expected from a heat-recovery boiler and thus vary with cycle pressure ratio and turbine inlet temperature. For a pressure ratio of 8 and a turbine-inlet temperature of 1400 K, the cycle efficiency improves from about 27.5 percent with no steam to 37.5 percent with steam at 23.6 percent of the air flow rate. The same steam-injection rate for an engine with a pressure ratio of 16 increases the cycle efficiency from 32 to 40 percent. Because of the additional mass flow

rate, steam injection also results in a significant increase in the specific power output.

Combustion studies of the effects of water or steam addition have generally been oriented towards NO_x reduction efforts (6 to 8, for example). These studies have been for both premixed and diffusion-flame combustors. Catalytic combustion has displayed clear emissions and stability advantages over other types of combustion (9 and 10, e.g.) with distillate fuels, but it has not been studied with steam injection. There are two effects of steam injection which were addressed by the present study of catalytic combustion. The first was the effect of steam on catalyst performance. To establish this effect, combustion efficiency and emissions were determined for a range of steam flow rates for a high-nitrogen fuel (an SRC II coal-derived liquid) and for a clean distillate fuel (no. 2 diesel). The second interest was in the determination of the effect of steam injection on the tendency for petroleum residual fuels to burn in the premixing duct upstream of the catalytic reactor. In the studies of (11 and 12) this upstream burning apparently occurred as the result of fuel being deposited on the duct walls, then igniting periodically. In studies for which combustor inlet-air temperatures were above 800 K, however, this problem disappeared and combustion of residual fuel was stable (11 and 13). To determine if steam injection might suppress this upstream burning, a petroleum residual fuel was tested with varying steam flow rates.

For these experiments, steam at a temperature of 450 K and at flow rates of 24 to 52 percent of the air flow rate was injected along with the test fuel to mix with the airstream upstream of the catalytic reactor. Steam and compressor-discharge-air temperatures for stationary gas-turbine engines will typically be in the range of 600 to 750 K. Because of the relatively low-temperature steam available for this study, inlet-air temperatures of 800 and 880 K were used to provide steam-air mixtures with temperatures in the range of 630 to

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740 K. All experiments were performed at a pressure of 600 kPa and an inlet-air velocity of 10 m/s.

DESCRIPTION OF EXPERIMENT

The steam-injection experiments were performed in the flame tube described in Fig. 3. It was lined internally with 12-cm-inside-diameter Carborundum T30R Fiberfrax tube insulation to minimize heat loss. The inlet air was indirectly heated to temperatures of 800 and 880 K. The inlet-air temperature was measured with an array of 12 Chromel-Alumel thermocouples located just upstream of the test section. Pressure was controlled to a constant value of 600 kPa for all tests with a back-pressure valve downstream of the test section shown in Fig. 3. The three test fuels, no. 2 diesel, petroleum residual, and SRC II middle-heavy blend, are described in Table 1. The residual-fuel lines were electrically heated to decrease the fuel viscosity sufficiently to permit it to be pumped.

Steam-Fuel Injection

Steam was supplied from a boiler at pressures of 1 to 1.4 MPa. The temperature at the test section was 450 K. Test-section fuel was mixed with the pressurized steam in 21 separate 0.635-cm-diameter tubes as shown in Fig. 3. No mechanical mixing devices were used; the fuel lines were simply connected to the steam lines with tee fittings. The injector was designed to distribute equal quantities of steam and equal quantities of fuel to each of 21 air passages. This was done by making all steam-fuel tubes the same length. However, preliminary tests with no steam flow resulted in the melting of the lower portion of the catalytic reactor. Apparently, the pressure drop in the steam-fuel tubes of the injector was insufficient for good distribution when fuel was flowing alone. As a result, the fuel-air ratio at the bottom of the duct was much richer than average. Thus, to insure uniform fuel-air ratio profiles, all tests were performed with a steam flow rate at least 24 percent of the air flow rate.

Each of the steam-fuel tubes was paired with one of the 21 diverging air passages which were machined in the fuel injector body. Figure 4(a) shows the pattern of these passages while Fig. 4(b) is a cutaway view. Each diffusing air passage continued until it had merged with its neighbors or the outer periphery of the injector body. By eliminating flow passage discontinuities, this design minimized the possibility of recirculation zones becoming established at the fuel injector exit.

Steam-Air Mixture Temperature

Steam flow rates were metered with an orifice. For each steam flow rate, steam-air mixture temperatures were measured without fuel flow with thermocouples mounted in the catalytic-reactor section. These measurements agreed closely with mixture temperatures calculated from steam and air temperatures and flow rates. Steam-air mixture temperatures reported here are the calculated values.

Steam-Fuel Air Mixture Temperature

The addition of liquid fuel to the steam will reduce the mixture temperature by 10 to 25 K below the reported steam-air mixture temperatures for the flow rates studied.

Detection of Upstream Burning

A single thermocouple was mounted in the premixing duct downstream of the fuel-steam injector. The signal from this thermocouple operated a relay to shut off fuel flow in the event of burning in the premixing duct.

Catalytic Reactor

The catalytic reactor was located 25 cm downstream of the plane of fuel injection. This distance permitted mixing of fuel, steam, and air. The catalytic reactor was designed to match configurations which have been successful in past catalytic combustion studies with coal-derived and residual fuels (11 to 15). These used a graded-cell catalyst; that is, large-cell catalyst elements were located at the inlet of the reactor to insure high surface temperatures (15) and avoid plugging by fuel deposits, and these were followed with elements having successively smaller cells to permit complete reaction of fuel and air. The graded-cell configuration used in the present study is described in Table II. Eight catalyst elements, 2.54 cm long and 12 cm in diameter were used. The first two elements had cells with a density of 3.9 cells/cm². These were followed with two elements with 15.5 cells/cm² and four elements with 46.5 cells/cm². Palladium catalyst was used on all elements. The catalyst elements were prevented from sliding downstream by thermocouples which were inserted radially into the duct between the elements.

Exhaust Gas Sampling

A single-point water-cooled gas-sampling probe was located 19.4 cm downstream of the catalytic reactor on the duct center-line. The exhaust-gas sample flowed continuously through a heated tube to on-line gas-analysis equipment. The analyzers included a flame-ionization detector for unburned hydrocarbons, non-dispersive infrared analyzers for CO and CO₂, and a chemiluminescent analyzer for total NO_x (NO + NO₂).

RESULTS AND DISCUSSION

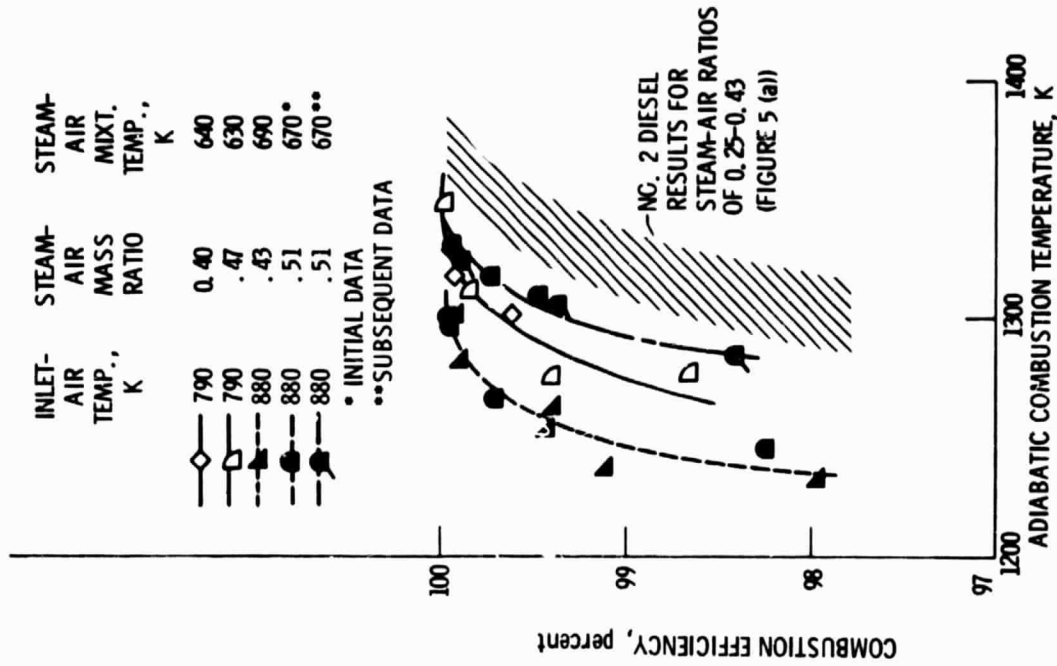
The velocity of the air as it approached the test section was 10 m/s and the pressure was 600 kPa for all tests. Results with the residual fuels will be discussed first, followed by a presentation of the combustion efficiency and emissions for the no. 2 diesel and SRC II fuels.

Residual Fuels

Experiments with residual fuel were performed with inlet-air temperatures of 800-880 K and with inlet steam-air mixture temperatures of 670-740 K. The catalytic reactions were started with no. 2 diesel fuel, then the residual was gradually substituted so that catalytic combustion was sustained totally with the residual fuel.

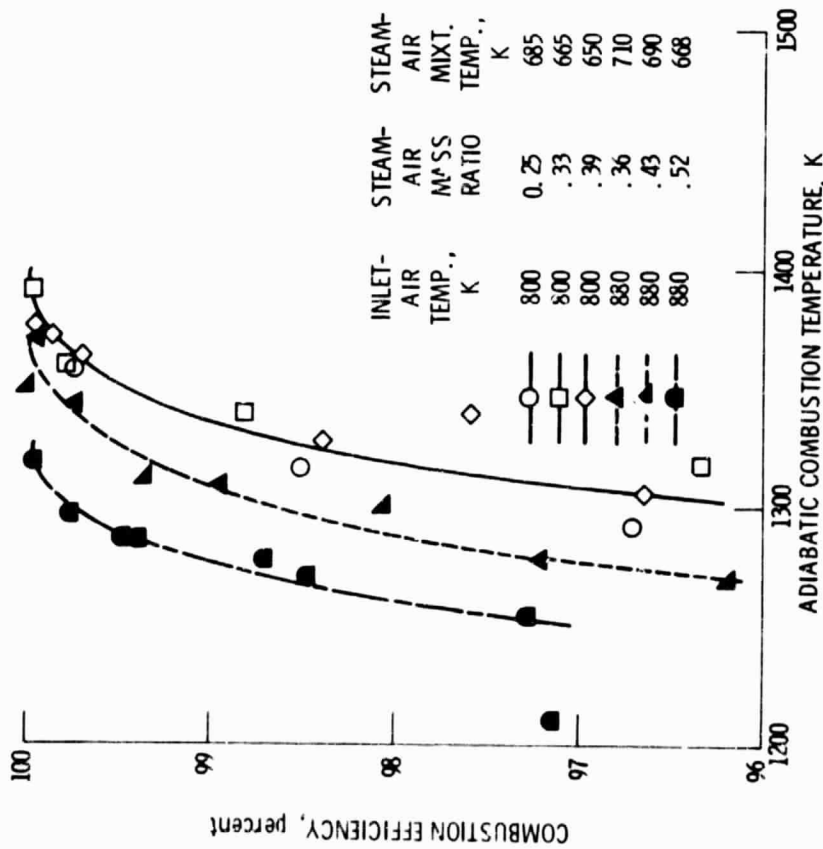
The main objective of the residual fuel testing was to determine if steam injection could be used to suppress upstream burning. While upstream burning was experienced with lower steam flow rates, increasing steam flow to above 30 percent of the air flow rate quenched the upstream reactions. Previous studies of catalytic combustion of residual fuel without steam injection experienced the coating of fuel injector surfaces, premixing duct walls, and catalytic reactor surfaces with fuel deposits at inlet-air temperatures below 800 K. Inspection of

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(b) Efficiency with SRC II mid-heavy blend.

Figure 5. - Concluded.



(a) Efficiency with no. 2 diesel fuel.

Figure 5. - Effect of combustion temperature on combustion efficiency. Inlet-air pressure, 600 kPa; inlet-air velocity, 10 m/s.

tion was observed when the steam-air ratio was increased to 0.52 at an inlet-air temperature of 880 K.

The carbon monoxide emissions for the mid-heavy blend of SRC II is given in Fig. 6(b). The trends seen in this figure are the same as those observed previously. Again, steam-air ratio had little effect on the results, and emissions could be reduced by increasing the inlet-air temperature. Emissions approached 1 g CO/kg fuel for combustion temperatures of 1300 to 1350 K. The unexpected drop in combustion efficiency for the case of a steam-air ratio of 0.51 (see discussion of Fig. 5(b)) can be seen from Fig. 6(b) to be the result of an increase in CO emissions.

Nitrogen Oxides Emissions

The effect of the adiabatic combustion temperature on NO_x emissions is presented in Fig. 7. Figure 7 gives the NO_x emission index as a function of adiabatic combustion temperature for both no. 2 diesel and SRC II mid-heavy blend. Because catalytic combustion provides high efficiencies at low combustion temperatures, virtually all of the NO_x produced is from the conversion of fuel-bound nitrogen into NO or NO_2 . In the case of the no. 2 diesel fuel, there is little fuel nitrogen so that NO_x emissions were low - typically between 0.2 and 0.25 g NO_2 /kg fuel. These values are well below the EPA new-source standard for stationary power plants of 6.3 g NO_2 /kg fuel.

For fuels having fuel nitrogen in excess of 0.25 percent, the new-source standard is 10.5 g NO_2 /kg fuel. NO_x emissions for the SRC II fuel increased with increasing adiabatic combustion temperature to values of 15 g NO_2 /kg fuel. For combustion temperatures high enough to insure high combustion efficiency (temperatures above 1300 K), NO_x emissions with the SRC II fuel were well above the 10.5-g NO_2 /kg fuel standard. NO_x emissions with both fuels were independent of steam-injection rates within the experimental accuracy.

Complete conversion of the 0.95 percent nitrogen in the SRC II fuel would result in an emission index of 31.2 g NO_2 /kg fuel. The no. 2 diesel is reported as having 0.014 percent nitrogen; complete conversion of this nitrogen into NO and NO_2 would yield an emission index of 0.46 g NO_2 /kg fuel. Because low concentrations of fuel nitrogen are difficult to measure accurately, fuel-nitrogen conversions for the no. 2 diesel fuel will not be presented.

The fuel-nitrogen conversion for the SRC II is given in Fig. 7(b) with the combustion efficiency as the abscissa to show the correlation between the two results. Within the experimental data scatter, fuel nitrogen conversion correlates well with combustion efficiency. As test conditions changed such that combustion efficiency increased, more and more of the fuel nitrogen reacted to form NO_x . For a combustion efficiency of 98 percent, only about 30 percent of the fuel nitrogen became NO_x . As combustion efficiency approached 100 percent, however, fuel-nitrogen conversion approached 50 percent. This result was independent of steam flow rate or inlet-air temperature. As the fuel reacts some fuel nitrogen is released to form N_2 and some remains in intermediate species such as HCN and NH_3 which were not measured in this study. Virtually all of the combustion inefficiency in this study was the result of higher-than-equilibrium CO concentrations, and very little unburned hydrocarbon was detected; thus the correlation could have been made equally well with CO emissions as with combustion effi-

ciency. Although residence time effects were not studied, the fact that N conversion correlates with combustion efficiency (CO), suggests that changes in residence time would have the same impact on both nitrogen conversion and combustion efficiency (CO). This observation leads to the conclusion that the intermediate nitrogen-containing species must oxidize at about the same rate as CO at these test conditions. With residence times higher than those of this study, nitrogen conversions in excess of 50 percent might occur. Such a residence-time effect on NO_x conversion was observed in the study of (14).

While the maximum conversion experienced in this study was well below the 70 percent reported for the same fuel in (14), it was still too high to meet the EPA new-source standard (see Fig. 7(a)). It is not likely that a simple single-stage catalytic combustor can provide low-emissions combustion of fuels containing high bound-nitrogen levels.

SUMMARY OF RESULTS

Catalytic combustion experiments were performed with steam injected along with fuel into the inlet airstream in a 12-cm diameter duct. A petroleum residual fuel, a mid-heavy SRC II blend, and no. 2 diesel fuel were tested at inlet-air temperatures of 800 and 880 K, an inlet-air velocity of 10 m/s, an inlet-air pressure of 600 kPa, and with steam flow rates of 24 to 52 percent of the air flow rate. Steam-air mixture temperatures were in the range of 630 to 740 K. The primary findings for these experiments can be summarized as follows:

1. For residual-fuel combustion, inlet steam-air mixture temperatures were not high enough to permit stable catalytic combustion; however, steam did prevent upstream burning from occurring if the steam flow rate was at least 30 percent of the air flow rate.
 2. Catalytic combustion of both the no. 2 diesel and SRC II was stable for all test conditions. Performance was not affected by increases in steam-air ratio except that the highest steam flow improved performance with no. 2 diesel fuel. For an inlet-air temperature of 880 K, a steam-air ratio of 0.52 and a steam-air mixture temperature of 668 K, the combustion efficiency was greater than 99.9 percent for combustion temperatures greater than 1310 K for no. 2 flow diesel or temperatures greater than 1290 K for the SRC II.
 3. The carbon monoxide emissions at an inlet-air temperature of 880 K, a steam-air ratio of 0.52, a steam-air mixture temperature of 668 K, and a combustion temperature of 1300 K were 10 g CO/kg fuel for the no. 2 diesel fuel and 1.5 g CO/kg fuel for the SRC II.
 4. NO_x emissions were in the range of 0.19 to 0.25 g NO_2 /kg fuel for the no. 2 diesel and 6 to 16 for the SRC II. These emissions increased with combustion temperature, but were independent of all other test variables. Conversion of fuel nitrogen to NO_x for the SRC II was shown to have a direct correlation with combustion efficiency. The conversion increased to a maximum of 50 percent as combustion efficiency approached 100 percent. For conditions at which complete combustion was achieved, the conversion of fuel nitrogen to NO_x was too great to permit meeting the EPA new-source standard with the SRC II fuel.
- This study showed that steam apparently acts only as a diluent when added to a catalytic combustion system. It has no adverse effect on combustion

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performance or emissions. The results suggested that for some conditions steam injection may improve performance, provably by improving fuel atomization and distribution.

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TABLE 1. - PROPERTIES OF TEST FUELS

	Residual	SRC 11 Mid-heavy blend	No. 2 diesel
Distillation, percent:	Temperature, K		
5	530		478
10	572	484	487
15	608		
20		495	500
50		522	533
70		546	553
90		627	581
FBP		644	596
Elements:	Percent by weight		
Carbon	87.16	86.21	86.7
Hydrogen	10.66	8.64	13.0
Nitrogen	0.53	0.95	0.014
Sulfur	0.16	0.21	0.19
Specific gravity at 289 K	0.899	0.999	0.848
Viscosity, cS	101 at 339 K 25 at 367	4.53 at 311 K -----	3.76 at 295 K -----
Gross heating value, 10 ⁷ J/kg	4.372	3.996	4.491

TABLE II. - CATALYTIC REACTOR DESCRIPTION

Element	Catalyst		Substrate		Cell density, cells/cm ²
	Material	Manufacturer	Material	Manufacturer	
1	Palladium	Retallick	Mullite	DuPont	3.9
2	↓	Retallick	↓	↓	3.9
3		Mex Pro			15.5
4		↓			15.5
5			Cordierite	Corning	46.5
6			↓	↓	
7					
8					

Each element is 2.54 cm long and 12 cm in diameter.

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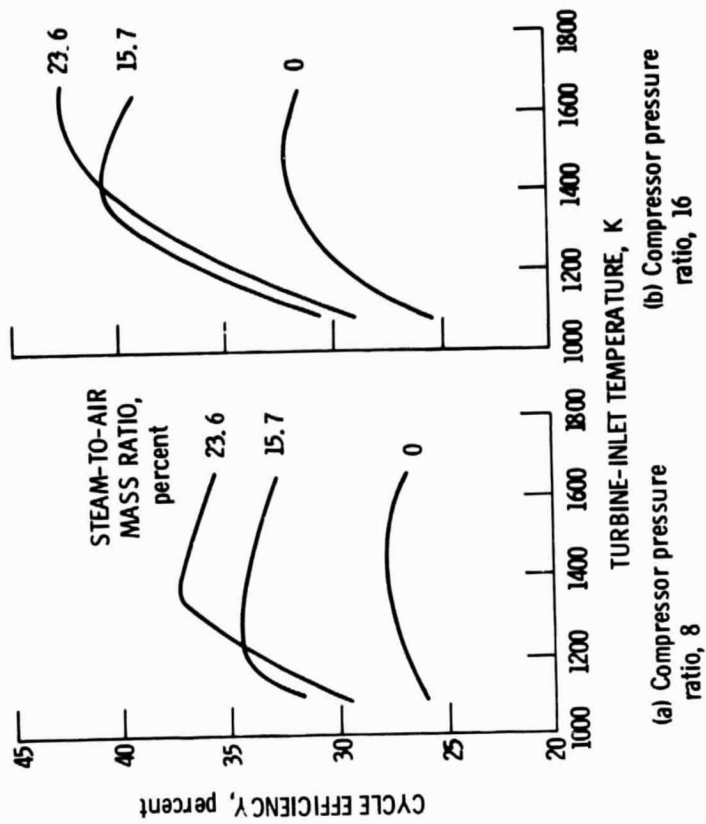


Figure 2. - Effect of steam injection on gas-turbine cycle efficiency
(from ref. 1).

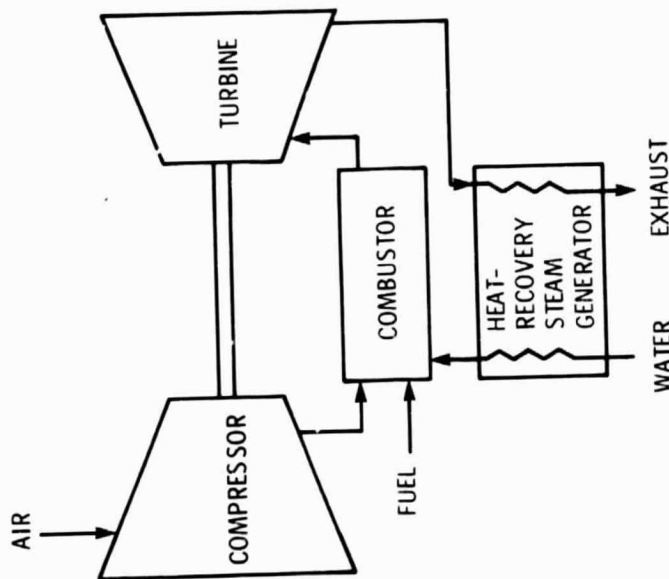


Figure 1. - Steam-injected gas-turbine.

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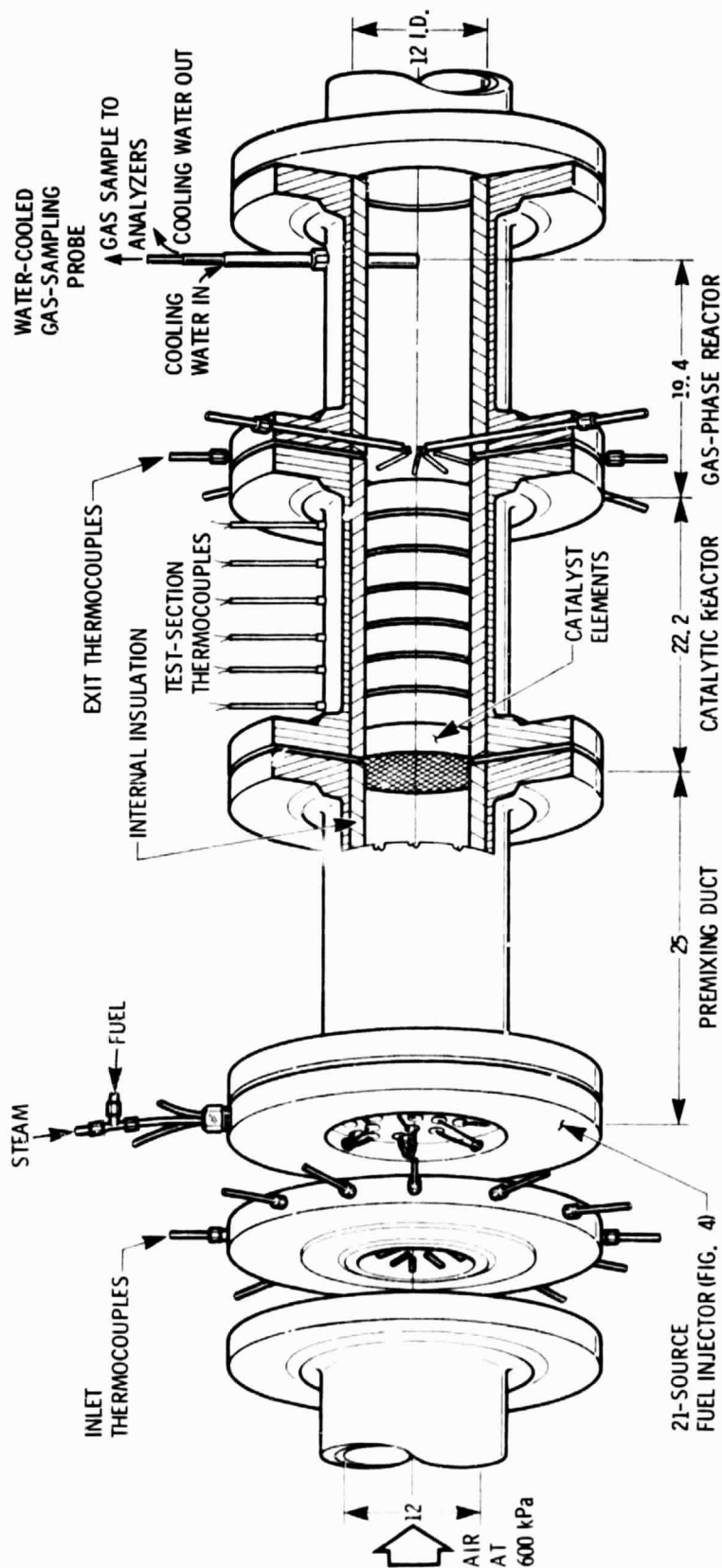
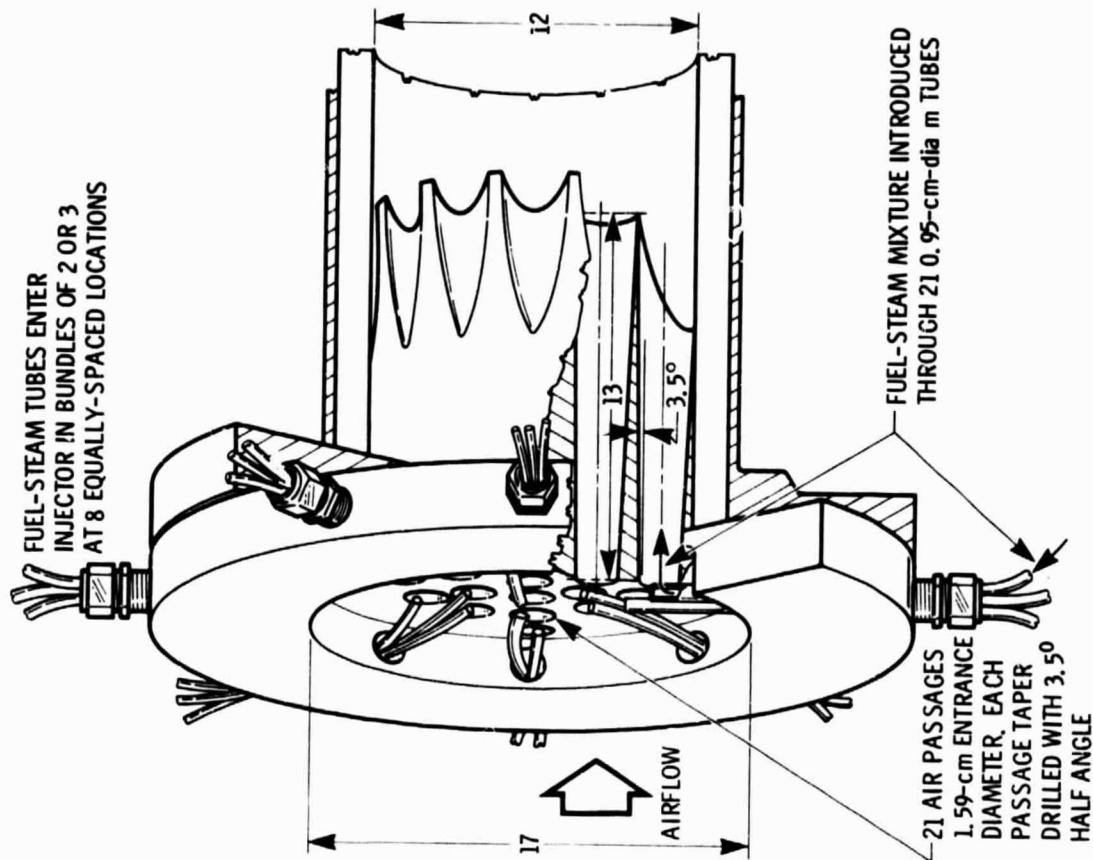


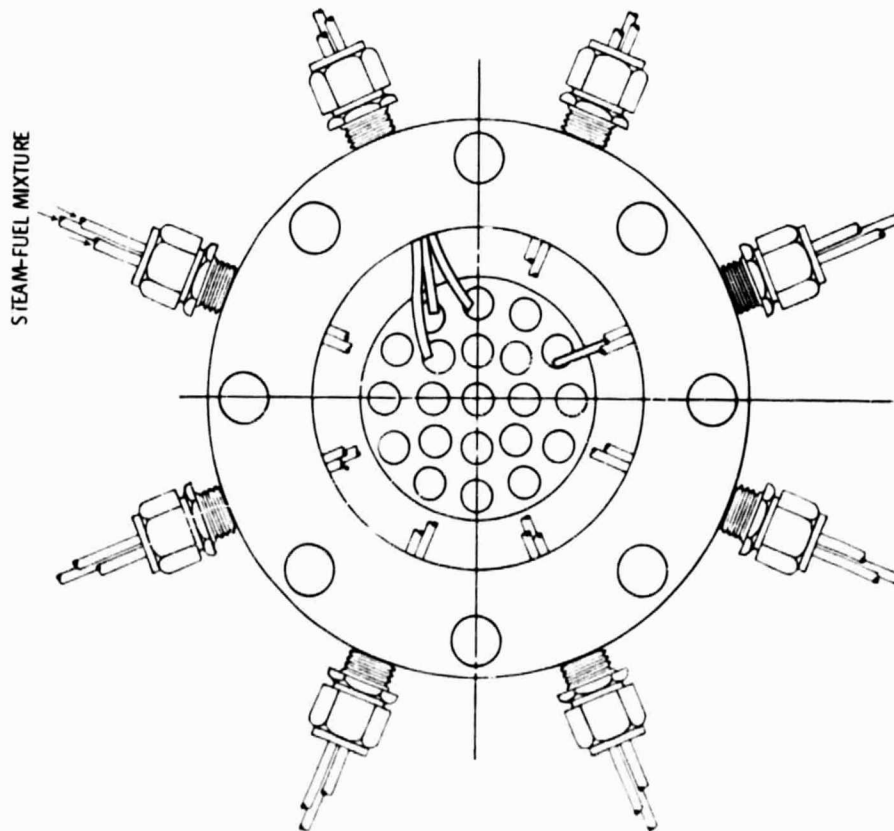
Figure 3. Test section. (Dimensions are in cm.)

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(b) Side view and cross section.

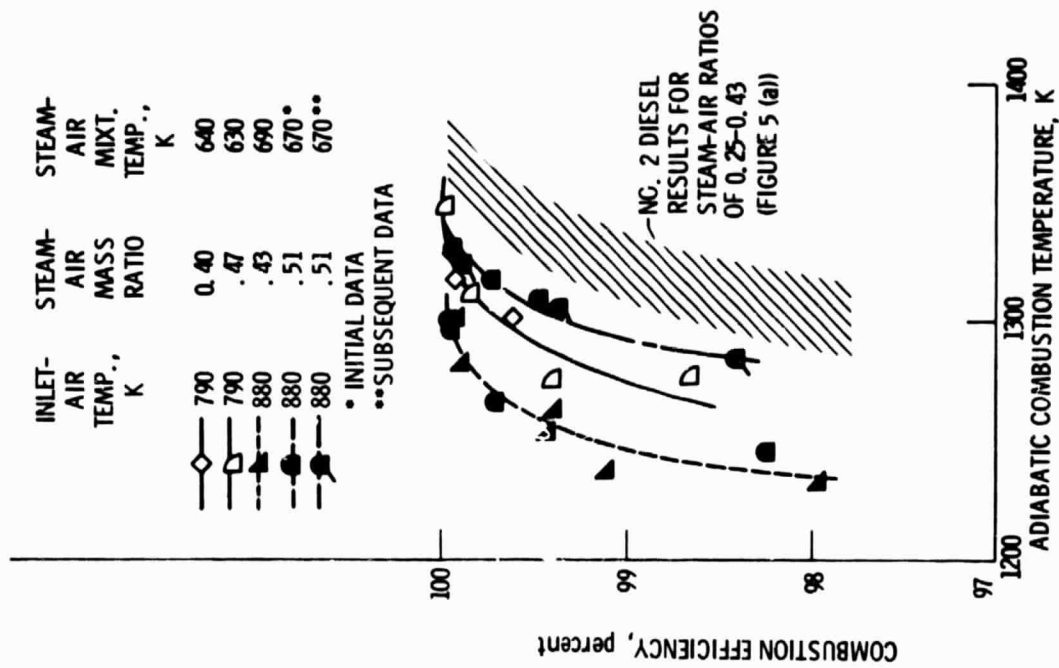
Figure 4. - Concluded.



(a) Front view showing pattern of air passages.

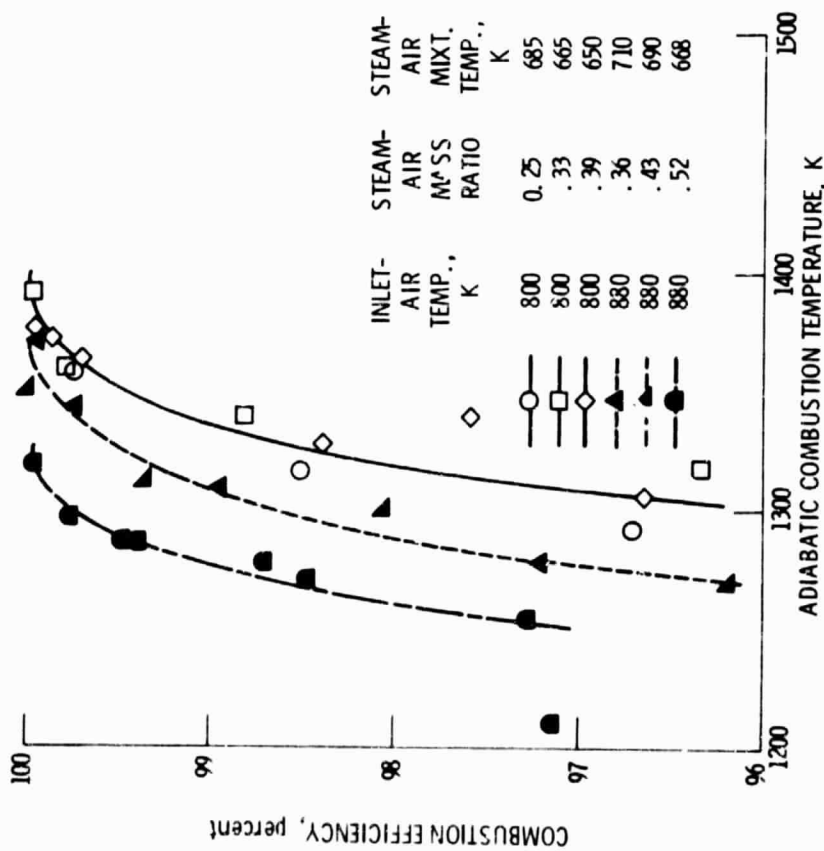
Figure 4. - Fuel-steam injector.

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(b) Efficiency with SRC II mid-heavy blend.

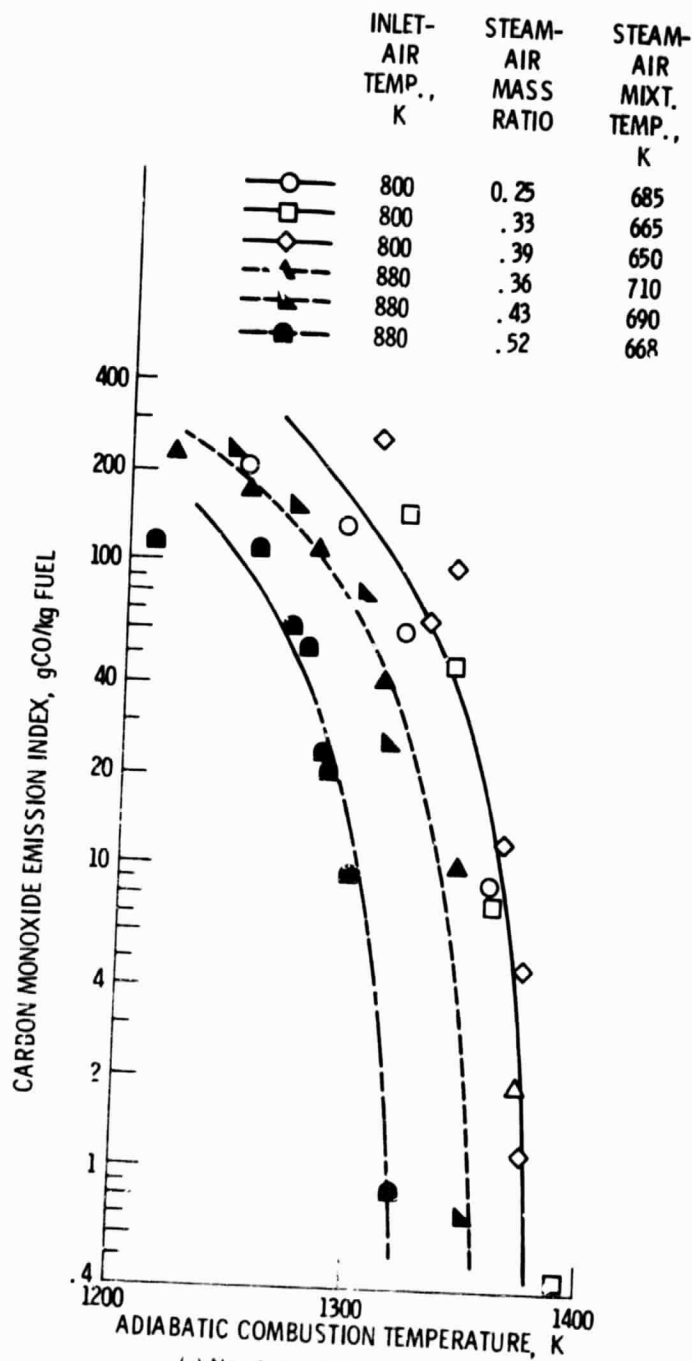
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(a) Efficiency with no. 2 diesel fuel.

Figure 5. - Effect of combustion temperature on combustion efficiency. Inlet-air pressure, 600 kPa; inlet-air velocity, 10 m/s.

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(a) No. 2 diesel fuel emissions.

Figure 6. - Effect of combustion temperature on carbon monoxide emissions. Inlet-air pressure, 600 kPa; inlet-air velocity, 10 m/s.

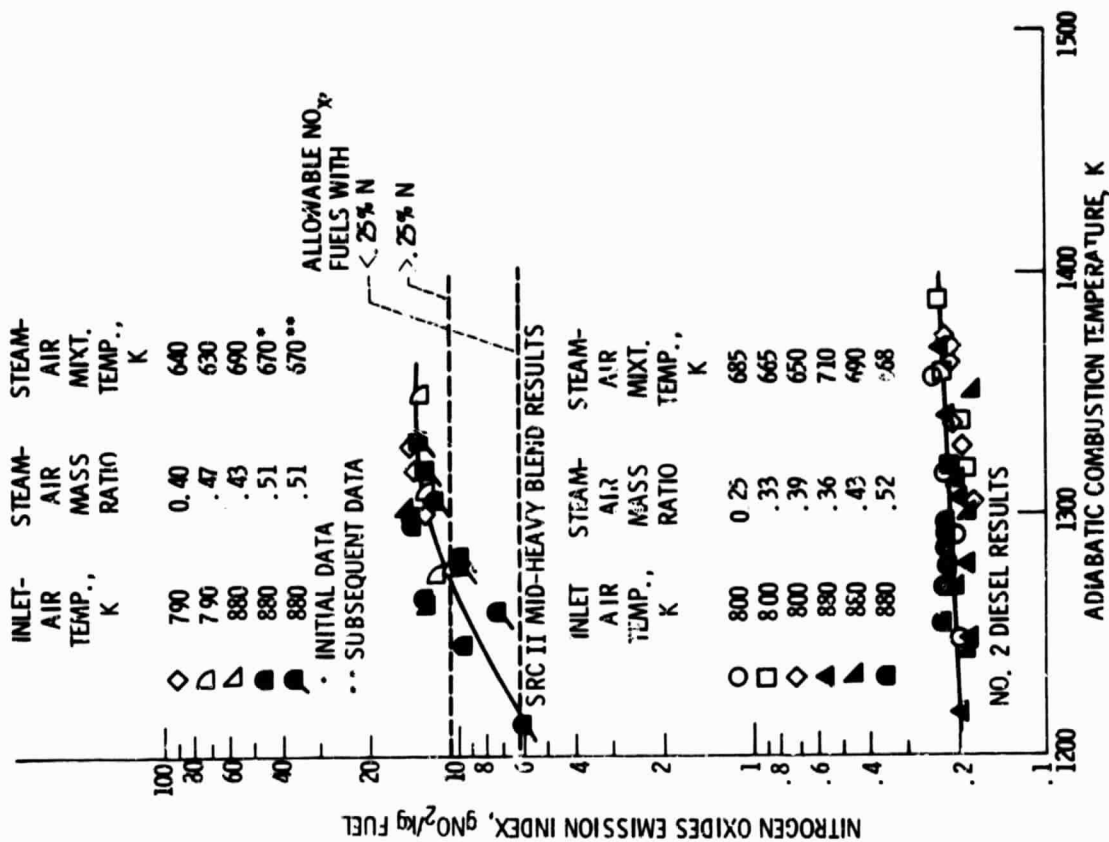
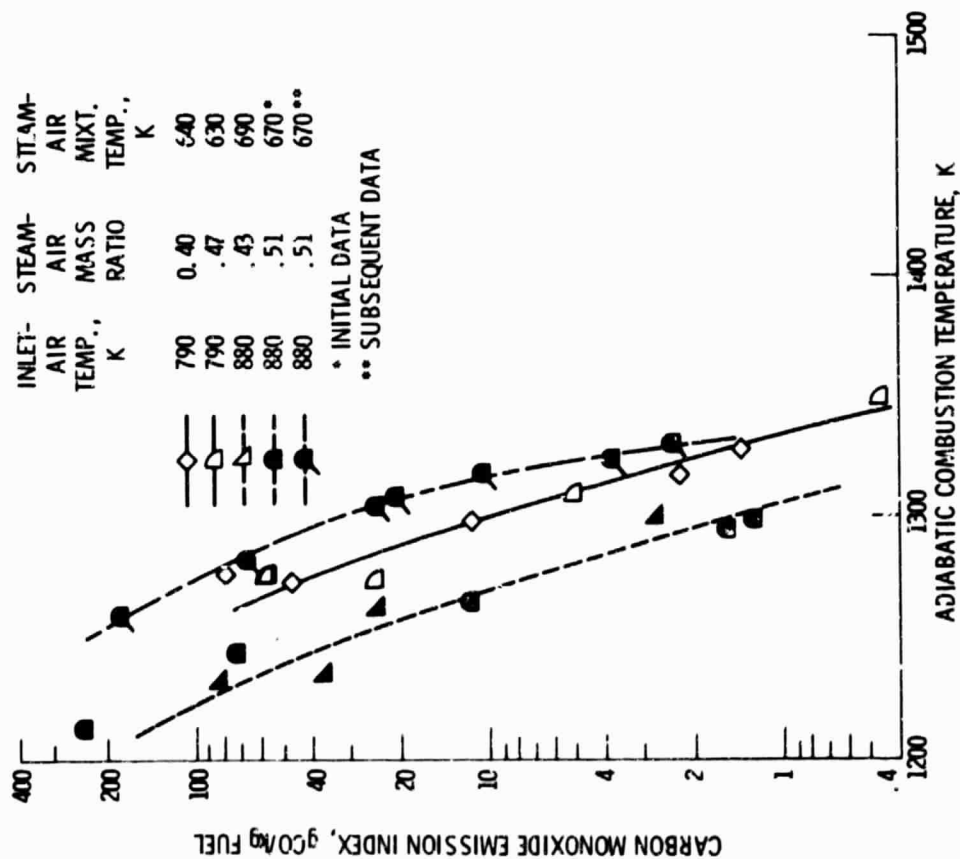


Figure 7. - Effect of combustion temperature on nitrogen oxides emissions. Inlet-air pressure, 600 kPa; inlet-air velocity, 10 m/s.



(b) SRC II mid-heavy blend emissions.

Figure 6. - Concluded.

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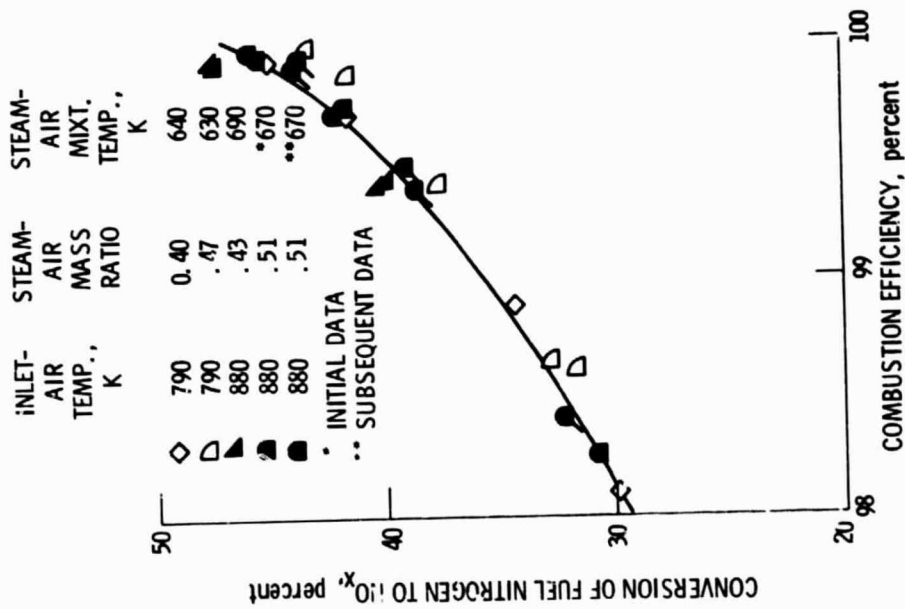


Figure 8. - Correlation of fuel nitrogen conversion with combustion efficiency for SRC II mid-heavy blend. Inlet-air pressure, 600 kPa; inlet-air velocity, 10 m/s.