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# Fuel-Rich Catalytic Combustion—A Fuel Processor for High-Speed Propulsion

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# FUEL-RICH CATALYTIC COMBUSTION - A FUEL PROCESSOR FOR HIGH-SPEED PROPULSION

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## INTRODUCTION

In a previous publication (ref. 1), results were presented for the fuel-rich catalytic combustion of iso-octane (equivalence ratios of 4.0 to 8.0). In addition to producing a very reactive and soot-free fuel with temperatures up to 1200 K, the process was shown to upgrade the structure of the fuel by the formation of hydrogen and smaller hydrocarbons. It also produced a cleaner burning fuel by removing some of the fuel carbon from the soot formation chain.

With fuel-rich catalytic combustion as the first stage of a two-stage combustion system, these enhanced fuel properties can be utilized by both high speed engines, where time for ignition and complete combustion is limited, and engines where emission of thermal  $\text{NO}_x$  is critical. Two-stage combustion has been shown to be effective for  $\text{NO}_x$  reduction in stationary burners where residence times are long enough to burn up any soot formed in the first stage. Such residence times are not available in aircraft engines. Thus, the soot-free nature of the present process is critical for high-speed engines. Hustal and Sonju (ref. 2) studied the lower flammability limit of hydrocarbons as a function of temperature and showed that the limit goes to zero at about 1473 K. It is estimated that the maximum temperature for the gases produced from fuel-rich catalytic combustion is probably 1350 K which is the threshold temperature for the formation of soot (ref. 3). Thus, the high temperature produced by this process enables one to use fuel-lean combustion in a second stage as a technique for controlling the combustion temperature and the formation of thermal  $\text{NO}_x$ .

Success of this concept will be highly dependent on the ability to apply it to practical and commonly used aircraft fuels, such as Jet-A and JP-4. This paper will describe the successful application of fuel-rich catalytic combustion to Jet-A, a multicomponent fuel used in gas turbine combustors. The further objective of ongoing investigations will be to then apply the concept to a staged combustor where approximately 6 to 8 percent of the engine airflow is diverted to the catalytic combustion (fuel preparation) stage and then utilizing a high-speed mixing device to mix the partially reacted gaseous fuel with the remainder of the combustion air. Since the temperature and residence time of the process can be well controlled, it is expected that the second stage of the combustion process will also be soot-free and produce a minimal amount of thermal  $\text{NO}_x$ .

The experimental results presented in this study were obtained from two test facilities; I, a small bench scale flow tube reactor (fig. 1) which had an airflow of about 23 kg/hr and II, a scaled up version (fig. 2) with an airflow rate of 900 to 1400 kg/hr.

#### TEST FACILITY I - EXPERIMENTAL APPARATUS AND PROCEDURES

The catalytic flow tube reactor has been described in reference 1 and the present fuel injector in reference 4. The fuel used for this work is specified by ASTM Jet-A turbine fuel designation. This is a multicomponent kerosene type fuel commonly used in gas turbine engines. Jet-A with a H/C ratio of 1.92 has an autoignition temperature of 533 K and contains 15 percent aromatic and 1 percent olefin hydrocarbons. The fuel was metered to the reactor from a pressurized fuel tank. Flow rates measured with a calibrated turbine flow meter varied from 150 to 220 cm<sup>3</sup>/min, depending upon the equivalence ratio desired.

Standard procedures were followed for each run. These included a warm-up of at least 90 min with hot nitrogen followed by an additional warm-up of 30 to 50 min at the desired test conditions. This procedure assured steady-state temperatures in the reactor.

After the reactor reached a steady-state temperature, start-up was initiated by adding fuel to the hot nitrogen. To ensure that the equivalence ratio in the reactor was always beyond the rich limit of combustion, the flow system was designed to permit switching the gas from nitrogen to air without interrupting the flow. By adjustment of the regulated pressure of the two gases, the fuel-air equivalence ratio could be established with hot nitrogen and then switched to air. In this way the catalyst never experienced stoichiometric burning. Shutdown was accomplished by reversing the process. A calibrated strain gauge flow meter measured the airflow which was kept constant at about 0.3 std m<sup>3</sup>/min. The pressure at the inlet to the catalyst bed was maintained at about 160 kPa.

The catalyst used in these experiments was a 2:1 mixture by weight of palladium/platinum applied to cordierite honeycomb substrates coated with alumina washcoat. Three catalyst configurations were used in the study.

Gas samples were drawn from the reactor from a probe located 17.8 cm downstream of the catalyst bed, corresponding to about 20 msec of gas phase reaction time. The sampling system and procedure has been previously described (ref. 1). The gaseous combustion products were analyzed by gas chromatography with thermal conductivity and flame ionization detectors. Three columns were used for the separation of the gas components: (1) washed Molecular sieve, (2) Spherocarb, and (3) Porapak R.

#### RESULTS

Jet-A fuel was studied over the equivalence ratio range 4.8 to 7.8 (F/A = 0.331 to 0.538) with combustion temperatures ranging from 1200 to 1060 K (2160 to 1900 °R). Flashback and spontaneous ignition of the fuel were not a problem for the test conditions used in the present study. In prior tests with

a different fuel injector, the fuel autoignited ahead of the catalyst and the gases observed through the windows 7.6 cm downstream of the catalyst were completely opaque indicating large amounts of soot. With a fuel injector designed to better atomize the fuel, autoignition did not occur and the reaction products were completely transparent. Thus the soot-free nature of the process was demonstrated by the observation of a completely transparent reaction product.

The reaction temperature was measured 3.8 cm downstream of the catalyst bed with a shielded closed-end thermocouple. Figure 3 is a plot of the measured reaction temperature versus equivalence ratio. These measured temperatures were compared with combustion temperatures calculated by assuming that soot was or was not a product. The equilibrium calculations were performed with the NASA Lewis Research Center equilibrium program (ref. 5), which contains thermodynamic data for about 140 C-H-O-N species. This large data base was important because the no-soot calculated reaction temperatures depend not only on the initial conditions of the reactants (equivalence ratio and temperature), but also on which species could be considered reaction products. The temperatures predicted by the two calculations (soot/no-soot) differ by about 150 K, and as can be seen, the measured reactor centerline temperature agreed well with the no-soot line. This is the same behavior observed in the study of the catalytic combustion of iso-octane (fig. 3) and shows that the no-soot calculation is a good method for predicting the combustion temperature.

Table I shows a comparison of the reaction products, temperatures, and equivalence ratios for one of the catalyst configurations used in this study. The products are divided into two groups to show the fuel-processing character of the preburner. Group 1 contains  $H_2$ ,  $O_2$ ,  $N_2$ , and all  $C_1$  and  $C_2$  carbon containing compounds while group 2 contains the larger hydrocarbons. The analysis shows that the breakdown of Jet-A is very similar to that found for iso-octane with the exception of hydrogen and aromatics. The smaller hydrogen concentrations is probably due to the lower hydrogen-to-carbon ratio of the Jet-A fuel. Although Jet-A had an initial aromatic content of about 15 percent, less than 1 percent benzene and toluene was measured in the reaction products.

The carbon-atom balance across the catalyst bed was calculated from the measured reaction products. Changes in the number of moles of gas due to chemical reaction were accounted for by using nitrogen as a reference gas and balancing the carbon-nitrogen ratio across the catalyst bed. One molecule of Jet-A was assumed to contain 12 carbon atoms. Figure 4 is a plot of the percentage of the initial fuel carbon found in the product gases as a function of the equivalence ratio. Unlike the iso-octane study, we were unable to account for all of the fuel carbon of Jet-A with the gaseous products, since some of the fuel carbon was condensed in the ice bath cold trap along with the water. Calculations show that the loss of as little as 0.5 percent of a  $C_9$  compound in the cold trap would account for the missing carbon. Three catalyst configurations were studied and they clearly show that excess catalyst does not improve the carbon atom balance.

The fuel processing character of the process can be seen in figure 5, where the fraction of the fuel carbon in  $C_1$ ,  $C_2$ , and  $C_3$  molecules is plotted as a function of the equivalence ratio. At an equivalence ratio of 4.6, about 80 percent of the fuel carbon is in  $C_1$ ,  $C_2$ , and  $C_3$  molecules. Excess catalyst has no effect on the amount of carbon in this group.

## TEST FACILITY II

Preliminary data from the larger reactor will be used to determine the catalyst loading requirements and the effect of higher inlet air temperatures. Test conditions; changing mass flow rates, inlet combustion air temperatures, and fuel-to-air equivalence ratio (E.R.), are run in a matrix test fashion.

The initial catalyst tested consisted of 15.2-cm diameter monolithic cell ceramic disks which had been coated with a proprietary platinum base catalyst material. The disks were 2.5 cm thick and four of them were closely stacked together in series in the test unit. The first two disks had 100 equal size cell passages per square inch of disk surface; the last two disks had 200 cells per square inch.

The catalyst unit was tested by flowing a known mixture of hot air and vaporized fuel through the catalyst disks until equilibrium temperature, pressure, and composition conditions were obtained. Generally this was after about 5 min of flow and the results reported herein are from the 4 to 5 min portion of such testing.

Gas temperatures before and after the catalyst bed were measured with sets of 12 circumferentially spaced thermocouples whose sensing tips were about 5 cm from the gas flow centerline. The thermocouple arrays were 10 cm upstream and downstream of the catalyst bed. At the 10-cm downstream location, a gas sampling probe was used to continuously withdraw a sample for chemical composition analysis.

Test results for 11 tests are presented in table II. Tests 1 to 5 were conducted at identical airflow rates and input temperatures over a range of (E.R.) from 4.7 to 7.0. The gas temperatures at the different measuring sites as well as the gas composition, in terms of percent are presented as a function of E.R. Nitric oxide concentrations were under 10 ppm over the entire E.R. range. Tests numbered 6 to 8 had the same input air temperature as 1 to 5, but 25 and 50 percent greater mass flow rates, respectively.

The variation of measured combustion gas temperatures as a function of changing equivalence ratios (E.R.) is presented in figure 6 along with the calculated lines (soot/no-soot). As can be seen the data are in agreement with the no-soot line for E.R.'s below 5.0 but depart significantly as the fuel mass flow (E.R.) is increased. This behavior suggests that the active surface area in the catalyst bed is not large enough to realize complete combustion. Experiments are being conducted with catalyst beds two to three times larger than that used in the present tests.

The constituents (hydrogen and carbon monoxide) in the combustion products from both facilities are presented in figure 7. Although insufficient catalyst surface area was indicated for the large reactor, the emission levels were relatively close in value to those measured with the small reactor. In addition they show the same trend as a function of equivalence ratio. A comparison of the measured combustion temperatures for the two rigs can be shown to be quite similar, suggesting that these species concentration are strongly dependent upon the reaction temperature rather than completeness of reaction. Additional testing is required to verify this suggestion.

Although we have not been able to duplicate test results from the small reactor, we have produced a high-temperature fuel containing large amounts of hydrogen and carbon monoxide. The balance of the fuel is completely vaporized and in various stages of pyrolysis and oxidation. Visual observations indicate that there is no soot present. Additional experiments are required to clarify the differences from the small reactor results.

#### SUMMARY OF RESULTS

Fuel-rich catalytic combustion of Jet-A fuel was studied over the equivalence ratio range 4.7 to 7.8, which yielded combustion temperatures of 1250 to 1060 K. The process was soot-free and the gaseous products were similar to those obtained in the iso-octane study. A carbon atom balance across the catalyst bed calculated for the gaseous products accounted for about 70 to 90 percent of the fuel carbon; the balance was condensed as a liquid in the cold trap. It was shown that 52 to 77 percent of the fuel carbon was  $C_1$ ,  $C_2$ , and  $C_3$  molecules. This study demonstrated the viability of using fuel-rich catalytic combustion as a technique for preheating a practical fuel to very high temperatures.

Preliminary results from the scaled up version of the catalytic combustor produced a high-temperature fuel containing large amounts of hydrogen and carbon monoxide. The balance of the fuel was completely vaporized and in various stages of pyrolysis and oxidation. Visual observations indicate that there was no soot present.

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TABLE I. - GASEOUS REACTION PRODUCTS

	Catalyst surface area - 2067 cm <sup>2</sup>								
	1228	1213	1202	1190	1166	1146	1124	1107	1083
Reaction temperature, K	1228	1213	1202	1190	1166	1146	1124	1107	1083
Equivalence ratio	4.60	4.76	4.91	5.02	5.25	5.68	6.16	6.52	7.07
Fuel flow, g mol/min	0.580	0.602	0.623	0.646	0.757	0.823	0.895	0.958	1.037
Airflow, g mol/min	10.68	10.73	10.76	10.89	12.22	12.28	12.32	12.46	12.44
Residence time, msec	20.6	20.4	20.8	20.5	19.8	20.2	20.3	21.0	22.0
Pressure, kPa									
Upstream	156.7	156.0	156.4	155.8	160.3	159.7	159.0	159.2	159.4
Downstream	156.4	155.8	156.2	155.7	159.7	159.3	158.8	159.2	158.5
ΔP across bed	0.3	0.2	0.2	0.1	0.6	0.4	0.2	0.0	0.9
Temperature, K									
Preheated air	799	799	800	800	800	801	801	803	803
Fuel	302	302	301	301	304	303	303	302	301
Group 1:									
Hydrogen	7.70	7.23	6.83	6.26	5.58	4.76	3.92	3.43	2.83
Oxygen	0.89	0.91	0.96	0.99	1.05	1.03	1.13	1.22	1.40
Nitrogen	59.76	58.85	59.68	59.05	59.90	60.43	59.80	61.68	63.15
Carbon monoxide	16.84	16.47	15.94	15.74	15.02	13.89	12.39	11.96	10.94
Carbon dioxide	2.21	2.18	2.20	2.27	2.29	2.61	2.93	3.23	3.49
Methane	5.34	5.34	5.41	5.32	4.91	4.58	4.03	3.71	3.32
Acetylene	1.134	1.000	0.877	0.731	0.360	0.237	0.151	0.124	0.076
Ethylene	5.34	5.81	6.53	6.56	7.02	7.57	7.37	7.61	6.76
Ethane	0.09	0.13	0.16	0.21	0.38	0.47	0.55	0.64	0.59
Total Group 1	99.30	97.92	98.59	97.13	96.51	95.58	92.27	93.60	92.56
Group 2:									
Propylene	0.17	0.25	0.32	0.47	1.23	1.77	2.23	2.44	-----
C <sub>3</sub> hydrocarbons	0.133	0.155	0.171	0.189	0.191	0.162	0.075	0.058	-----
C <sub>4</sub> hydrocarbons	0.17	0.21	0.25	0.32	0.74	1.14	1.45	1.59	-----
C <sub>5</sub> hydrocarbons	0.04	0.05	0.06	0.07	0.16	0.28	0.44	0.56	-----
C <sub>6</sub> hydrocarbons	0.002	0.003	0.003	0.000	0.004	0.026	0.066	0.11	-----
Benzene	0.48	0.47	0.53	0.50	0.43	0.42	0.38	0.34	-----
Toluene	0.22	0.23	0.31	0.29	0.27	0.31	0.31	0.32	-----
C <sub>7</sub> hydrocarbons	0.000	0.000	0.000	0.000	0.000	0.005	0.015	0.027	-----
Styrene	0.040	0.040	0.063	0.043	0.030	0.011	0.017	0.011	-----
C <sub>8</sub> hydrocarbons	0.021	0.023	0.033	0.025	0.038	0.029	0.056	0.061	-----
Total Group 2	1.28	1.43	1.74	1.91	3.09	4.15	5.04	5.52	-----
Total Groups 1 and 2	100.58	99.35	100.33	99.04	99.60	99.73	97.31	99.12	-----

TABLE II. - PRELIMINARY TEST RESULTS  
[Platinum based catalyst test unit; Jet-A.]

Parameters	Test number										
	1	2	3	4	5	6	7	7	9	10	11
Equivalence ratio	4.7	4.9	5.7	6.7	7.0	4.4	5.2	5.8	4.5	4.9	5.2
Reaction temperature, K	1248	1194	1137	1074	1044	1239	1134	1074	1181	1112	1047
Combustion air											
Flow rate, kgm/hr	949	989	969	966	994	1175	1137	1169	1369	1378	1358
Temperature, K	947	943	946	944	945	936	937	939	909	908	910
Combustion products, vol. %											
Hydrogen	8.2	7.0	5.7	3.6	2.8	8.3	6.1	4.4	7.0	3.5	2.3
Carbon monoxide	13.9	13.4	11.2	8.8	8.0	11.9	12.7	10.7	13.1	9.1	6.3
Carbon dioxide	2.5	3.1	3.4	4.0	4.2	3.8	4.4	5.2	4.1	3.6	3.2
Oxygen	0.8	0.8	1.2	2.3	3.1	-----	-----	-----	-----	2.4	4.3



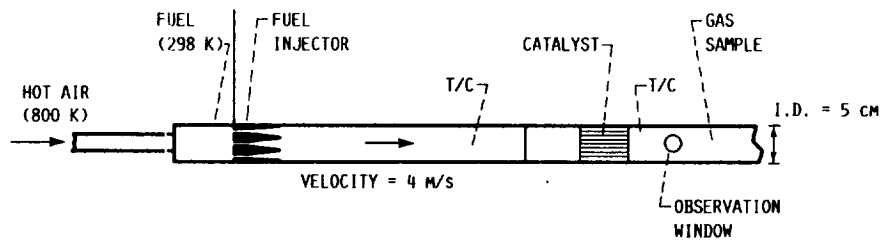


FIGURE 1. - SMALL BENCH SCALE APPARATUS (TEST FACILITY I) WITH AN AIR MASS FLOW OF 23 kg/HR.

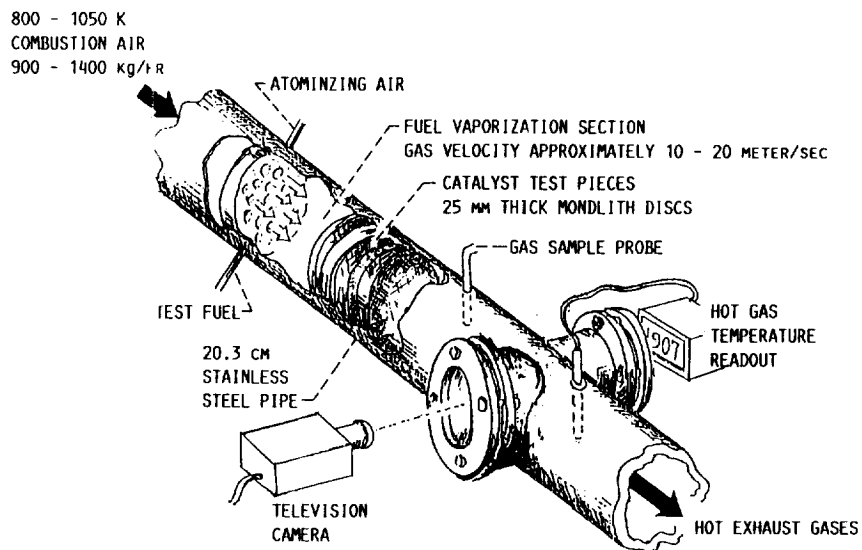


FIGURE 2. - LARGE TEST FACILITY II WITH AIR MASS FLOWRATES OF 900 - 1400 kg/HR.

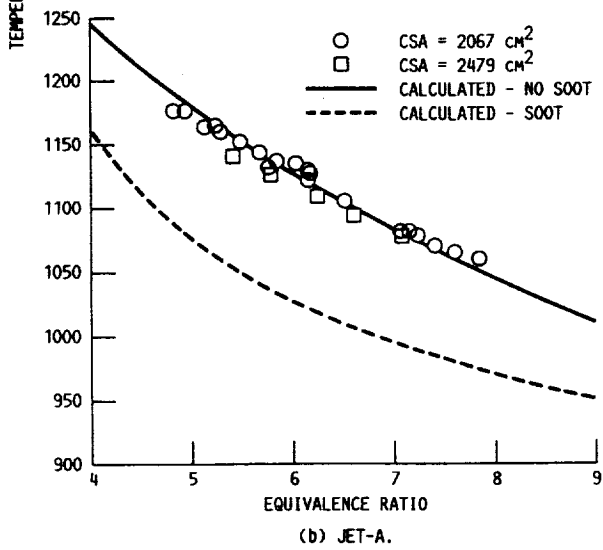
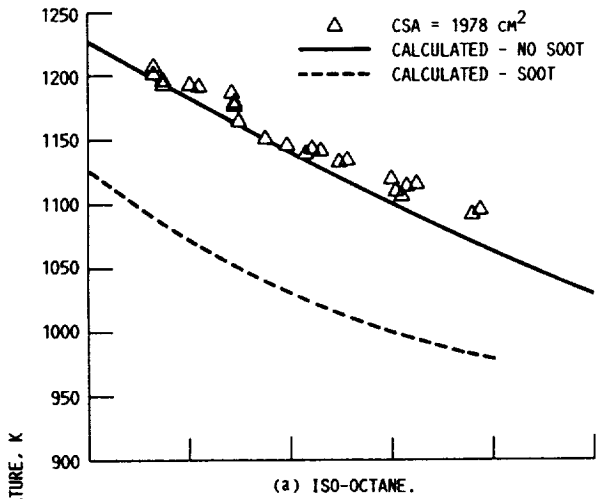


FIGURE 3. - COMPARISON OF MEASURED REACTION TEMPERATURES WITH CALCULATED EQUILIBRIUM TEMPERATURES (WITH AND WITHOUT SOOT) FOR ISO-OCTANE AND JET-A.

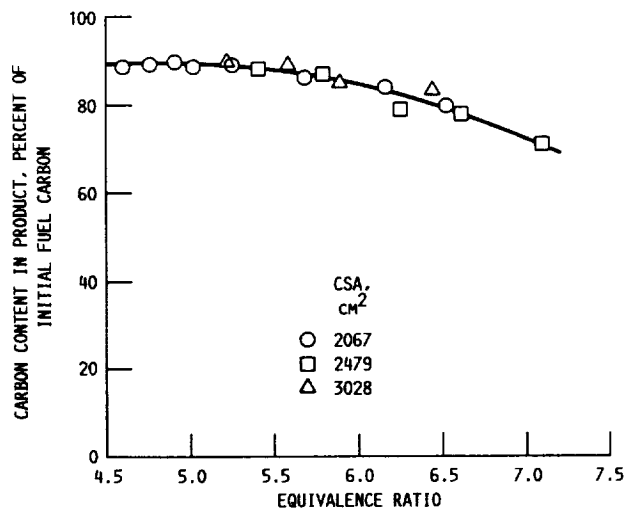


FIGURE 4. - PERCENT OF FUEL CARBON FOUND IN GASEOUS SAMPLES AS A FUNCTION OF EQUIVALENCE RATIO.

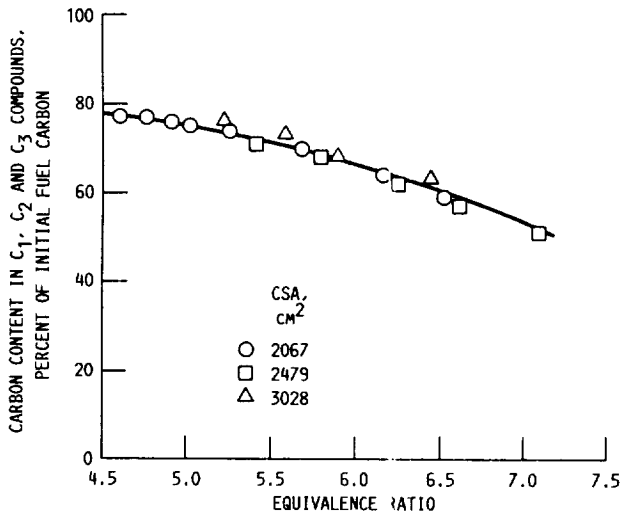


FIGURE 5. - PERCENT OF FUEL CARBON FOUND IN C<sub>1</sub>, C<sub>2</sub> AND C<sub>3</sub> COMPOUNDS AS A FUNCTION OF EQUIVALENCE RATIO.

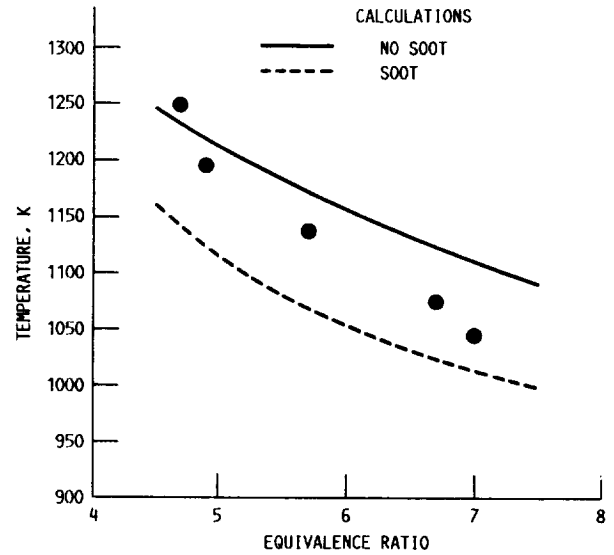


FIGURE 6. - COMPARISON OF MEASURED REACTION TEMPERATURES WITH CALCULATED EQUILIBRIUM TEMPERATURES AS A FUNCTION OF EQUIVALENCE RATIO IN TEST FACILITY II.

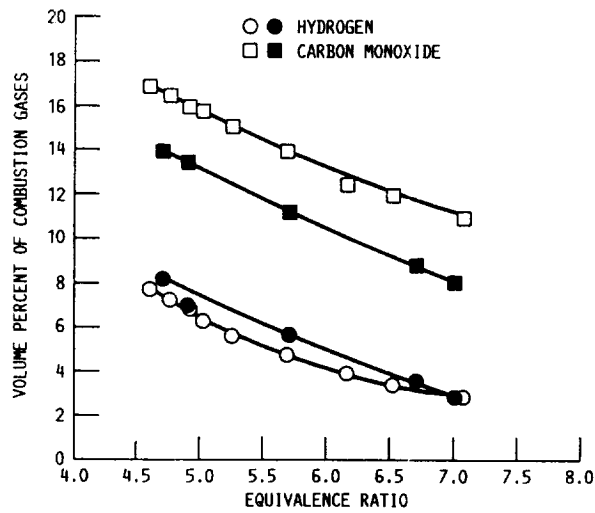


FIGURE 7. - COMPARISON OF THE EMISSIONS (HYDROGEN AND CARBON MONOXIDE) FOR THE TWO TEST FACILITIES. OPEN SYMBOLS ARE FOR TEST FACILITY I AND CLOSED ONES ARE FOR TEST FACILITY II.



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