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Catalytic Combustion of Coal-Derived Liquids

Daniel L. Bulzan and Robert R. Tacina
National Aeronautics and Space Administration
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U.S. DEPARTMENT OF ENERGY
Fossil Energy
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Twenty-sixth Annual International Gas Turbine Conference
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ABSTRACT
A noble metal catalytic reactor was tested with three grades of SRC 11 coal-derived liquids, naphtha, middle distillate, and a blend of three parts middle distillate to one part heavy distillate. A petroleum-derived number 2 diesel fuel was also tested to provide a direct comparison. The catalytic reactor was tested at inlet temperatures from 600 to 800 K, reference velocities from 10 to 20 m/s, lean fuel-air ratios, and a pressure of 3x10^5 Pa. Compared to the diesel, the naphtha gave slightly better combustion efficiency, the middle distillate was almost identical, and the middle-heavy blend was slightly poorer. The coal-derived liquid fuels contained from 0.58 to 0.95 % nitrogen by weight. Conversion of fuel nitrogen to NOX was approximately 75 % for all three grades of the coal-derived liquids.

INTRODUCTION
An experimental study was conducted to demonstrate catalytic combustion of coal-derived liquids obtained from the SRC II process and also to determine some of the potential problems associated with fuel preparation and catalytic combustion of the fuels. Petroleum-derived diesel fuel was also tested to provide a direct comparison of reactor performance with the coal-derived fuels.

Lewis Research Center is currently evaluating catalytic combustion as part of the Critical Research and Advanced Technology Support Project sponsored by the DOE Office of Fossil Energy, Division of Coal Utilization. One of the objectives of this program is to develop combustor concepts for stationary gas turbines which will fire coal-derived liquid fuels in an environmentally acceptable manner. Coal-derived fuels typically contain high levels of nitrogen which are readily converted to NOX during the combustion process. Coal-derived fuels have been tested in boilers (refs. 1 and 2) and gas turbine burners (ref. 3). Catalytic combustion of two coal-derived liquids, SRC II and H-Coal, is reported in reference 4. Catalytic combustion has demonstrated extremely low levels of thermal NOX operation (refs. 5 and 6); however, it has also shown high conversions of fuel bound nitrogen to NOX. Reference 4 reported essentially 100 % conversion of fuel bound nitrogen to NOX for lean fuel-air ratios. Additional tests were needed to help determine if catalytic combustion offers potential benefits for the combustion of coal-derived liquids.

A noble metal catalytic reactor was tested at inlet temperatures up to 800 K, reference velocities from 10 to 20 m/s, and a pressure of 3x10^5 Pa. Three grades of SRC 11 coal-derived liquids, naphtha, middle distillate, and a mid-heavy distillate blend, along with petroleum-based number 2 diesel fuel were tested. Performance and emissions of CO, CO2, NOX, and unburned hydrocarbons were measured.

EXPERIMENTAL DETAILS
Test Rig
The test rig is shown in Fig. 1. It was fabricated from 15.2-cm (6-in.-nominal-) diameter stainless steel pipe. Carborundum T30R fiberfrax tube insulation with a 12-cm inside diameter was inserted inside the pipe to minimize heat losses. A 0.15-cm thick stainless-steel liner was inserted inside the insulation upstream and downstream of the catalytic reactor to prevent erosion of the insulation. The inlet air was indirectly preheated and temperatures were measured at a plane just upstream of the fuel injector with an array of 12 Chromel-Alumel thermocouples mounted in a flange. Test section inlet pressure was measured at a tap located in the flange containing the inlet thermocouples. Pressure was controlled by a back pressure valve. The airflow
entering the test rig was measured by a standard ASME orifice. Fuel flow rates were measured using linear mass flowmeters.

Two types of multiple point fuel injectors were used. The first was an airblast type while the second was an air-assist type. A module of the hexagonal tube airblast fuel injector is shown in Fig. 2(a). Nineteen modules were welded together to comprise the complete fuel injector. Fuel was injected through a 0.07-cm inside diameter tube pointing downstream in the center of the smallest cross-sectional area of each module to provide good atomization and mixing. All fuel tubes were the same length, 25.4 cm, to provide equal flowrates to each module.

It was found the coal-derived liquids showed a tendency to plug the fuel tubes at elevated inlet air temperatures. As illustrated in Fig. 2(a), each fuel tube was surrounded by another tube to form a concentric tube arrangement. Air flowed through the 0.028-cm annulus to cool the fuel which prevented plugging of the fuel tubes. All testing was performed with cooling air flowing through the annulus. A module of the air-assist fuel injector is illustrated in Fig. 2(b). Fuel was injected downstream through the center, 0.024-cm inside diameter, tube. Air flowed through the outer ring of four, 0.031-cm inside diameter, tubes to assist in atomization and mixing of the fuel. It also provided cooling of the fuel tubes. Nineteen modules were used for the fuel injector. It was constructed such that the air-assist module arrangement was the same as the hexagonal tube module arrangement. The hexagonal tube modules, which made up the hexagonal tube fuel injector, were placed immediately upstream of the air-assist fuel injector to straighten the inlet velocity profile. A photograph of the air-assist fuel injector is shown in Fig. 3.

The premixing zone length of 30.6 cm was used for all fuels and fuel injectors. A single Chromel-Alumel thermocouple was used to detect any burning in the premixing zone. None was observed for these tests. The pressure drop across the fuel injector was measured with a differential pressure transducer connected between the inlet pressure tap and one located at the premixing region thermocouple station.

Two catalytic reactors were used for this study. The first was a uniform cell type while the second was a graded cell type. Both consisted of eight elements, 12 cm in diameter and 2.54 cm long, separated by a 0.31-cm gap containing at least one thermocouple. The last two elements were not separated. This arrangement is shown in Fig. 1. The reactor elements for both reactors are described in Table 1. The pressure drop across the catalytic reactor was measured with a differential pressure transducer connected between a tap at the premixing zone thermocouple station and one located in the flange at the first row of thermocouples downstream of the catalytic reactor.

At a distance 17.2 cm downstream of the catalytic reactor, a single point water-cooled gas sampling probe, 0.6 cm inside diameter sampling passage, was used to withdraw samples for emissions measurements. Temperatures were also measured downstream of the reactor at the axial locations shown in Fig. 1. The gas sample line was electrically heated to keep unburned hydrocarbons from condensing. Concentrations of CO and CO₂ were measured with nondispersive infrared analyzers, unburned hydrocarbons with a flame ionization detector, and nitrogen oxides (total NO + NO₂) with a chemiluminescent analyzer.

**Measurements and Computations**

**Reference Velocity**

The reference velocity was computed from the measured mass flow rate, the average inlet air temperature, the duct cross-sectional area, and the test-section inlet pressure.

**Emission Index**

Emissions were measured as concentrations in ppm by volume, corrected for water of combustion, and converted to emission indices using the expressions in reference 7.

**Combustion Efficiency**

Combustion efficiency was calculated from the following expression:

\[
\text{EFF} = 100 - 0.1 \left( \frac{\text{E.I.}_\text{HC}}{\text{E.I.}_\text{HC},\text{EQ}} - \frac{\text{HV}}{\text{HV}_\text{CO}} \left( \frac{\text{E.I.}_\text{CO}}{\text{E.I.}_\text{CO},\text{EQ}} - \frac{\text{E.I.}_\text{NO}_x,\text{EQ}}{\text{E.I.}_\text{NO}_x,\text{EQ}} \right) \right)
\]

where

- EFF = combustion efficiency, %
- E.I.₆g,₆Q = emission index of species X, g NO/avg fuel
- HV = lower heating value of X, J/kg
- E.I.₆g = emission index of species X, g NO/avg fuel
- CO = CO₂, and unburned hydrocarbons. The two values generally agreed within ±15 % of the adiabatic reaction temperature was computed from the computer program of reference 8 using the carbon balance fuel-air ratio. The carbon balance fuel-air ratio had the advantage that it was the local fuel-air ratio at which the emissions data were obtained. It also included the small amount of air (less than 3 % of the total airflow) used for cooling the fuel tubes.

**RESULTS AND DISCUSSION**

Three grades of SRC coal-derived liquids and petroleum-derived number 2 diesel fuel were tested. The properties of the fuels are listed in Table 11. As illustrated by the distillation curve values, the naphtha was the lightest grade tested and the mid-heavy blend (a blend of 2.9 to 1 of middle to heavy distillate) was the heaviest. Fuel nitrogen content of the coal-derived fuels ranged from 0.58 % for the naphtha to 0.95 % by weight for the mid-heavy blend. Hydrogen content varied from 12.09 % for the naphtha to 8.6 % by weight for the mid-heavy blend. Typical number 2 diesel fuel properties are listed for comparison. The testing sequence, along with the fuel injector and reactor used, is listed in Table III. All data presented were taken with the graded cell reactor of Table 1(b) because problems developed with the uniform cell reactor, which will be discussed later.

**Combustion Efficiency**

Combustion efficiency for all fuels is presented in Fig. 4. Combustion efficiency at an inlet temper-
The effect of inlet temperature and reference velocity on the unburned hydrocarbons emission index are shown in Figs. 11 and 12. Little effect is evident.

**NO\textsubscript{x} Emissions**

The NO\textsubscript{x} (sum of NO + NO\textsubscript{2}) emission index, expressed as g NO\textsubscript{x}/kg fuel, for all the fuels tested at three inlet temperatures is presented in Fig. 13. As expected, NO\textsubscript{x} emissions for the coal-derived liquids were considerably higher than the diesel fuel, due to the fuel bound nitrogen. The NO\textsubscript{x} emission index for the mid-heavy blend was about 105 ppm at 15% excess O\textsubscript{2}. Little effect is evident for reference velocity on NO\textsubscript{x} emissions. The nitrogen oxides standard for new, modified, and reconstructed stationary gas turbines is 125 ppm at 15% excess O\textsubscript{2} from reference 10. Fig. 14 shows the effect of fuel, and Fig. 15 shows the effect of inlet temperature on NO\textsubscript{x} emissions. The conversion of fuel nitrogen in percent to NO\textsubscript{x} is presented in Fig. 16, 17, and 18. Since thermal NO\textsubscript{x} emissions are negligible at relatively low temperatures of catalytic combustion (refs. 5 and 6), all measured NO\textsubscript{x} was assumed to originate from fuel nitrogen. The solid symbols indicate combustion efficiencies greater than 99%.

Fig. 16 presents the conversion in percent of fuel nitrogen to NO\textsubscript{x} for the three coal-derived fuels at inlet temperatures of 700 and 800 K. For both inlet temperatures, conversion increased rapidly with adiabatic reaction temperature and then leveled off. The naphtha gave higher conversions to NO\textsubscript{x} than the other tests on diesel, run A, run B, and run C, was essentially identical. The slightly better combustion efficiency of run A with diesel fuel, seen previously, was due to lower CO emissions.
bratic reaction temperature, the higher inlet temperature gave a higher conversion; however, the data for all three inlet temperatures approached a maximum conversion of about 75 %.

Fig. 18 illustrates the effect of reference velocity (residence time) on the percent conversion of fuel nitrogen to NOx at an inlet temperature of 800 K. Conversion approached 75 % at a reference velocity of 15 m/s, and about 55 % at 20 m/s. Since lower reference velocities gave an increased residence time in the catalytic reactor, increasing the catalytic bed residence time increases the conversion of fuel nitrogen to NOx.

Pressure Drop

The pressure drop as a percentage of the inlet pressure is presented in Fig. 19 for middle distillate fuel at reference velocities of 10, 15, and 20 m/s. At an adiabatic reaction temperature of 1350 K, the pressure drop ranged from 1.6 % at 10 m/s to 3.9 % at 20 m/s. This is reasonable for an application.

Other Results

Figures 20(a) and (b) show the reactor and fuel injector after operating on the mid-heavy blend at inlet-temperatures of 650 K. A deposit of about 0.3 cm thickness was formed on the front edges of the first reactor element. The fuel injector had deposits which were formed on each fuel injector module. Fig. 20(c) shows a portion of the first reactor element after it was operated at an inlet temperature of 600 K. As previously discussed, the front element quickly plugged at the lower inlet temperature which probably was caused by the decreased vaporization of the mid-heavy blend at 600 K.

A photograph of the uniform cell reactor, described in Table 1(a), is presented in Fig. 21 after it was operated at an inlet temperature of 600 K on the naphtha grade fuel. The reactor quickly plugged after the naphtha fuel was introduced. No further tests were made using the uniform cell reactor. Reference 11 reported decreased wall temperatures for the front elements of a uniform cell reactor as opposed to a graded cell type. It was found that the first element required a considerably longer length to increase from the inlet temperature to the adiabatic reaction temperature for the uniform cell reactor. This is probably the cause for the plugging of the uniform cell reactor on the naphtha fuel while the graded cell reactor operated on the fuel without any problems.

SUMMARY OF RESULTS

This study has demonstrated catalytic combustion of three grades of coal-derived liquids obtained from the SCR II process. The three grades tested were a naphtha, a middle distillate, and a mid-heavy distillate blend. Petroleum-derived diesel fuel was also tested to provide a direct comparison of catalytic reactor performance with the coal-derived fuel. Compared to the diesel, the naphtha was better than the diesel in combustion efficiency, the middle distillate was approximately equal to, and the mid-heavy blend was poorer than the diesel. The mid-heavy blend required inlet temperatures of at least 650 K to prevent plugging the channels of the first catalytic element.

The coal-derived fuels contained from 0.58 to 0.95 % of nitrogen by weight. Under the fuel lean conditions tested, catalytic combustion was capable of producing negligible thermal NOx emissions, was an efficient converter of fuel bound nitrogen to NOx. Conversion levels ranged as high as 75 %. The emission standard of 125 ppm NOx at 15 % excess O2 could not be met with any of the coal-derived fuels tested.

The catalytic reactor was operated for about 10 hours on the synthetic fuels. While no significant loss of activity or poisoning of the catalyst was apparent for the conditions tested, the application of catalytic combustion to stationary gas turbines would require operation for thousands of hours.

REFERENCES

### TABLE I. DESCRIPTION OF CATALYTIC REACTOR ELEMENTS

(a) Uniform cell reactor

<table>
<thead>
<tr>
<th>Element</th>
<th>Catalyst material</th>
<th>Catalyst loading, kg/m³</th>
<th>Substrate manufacturer</th>
<th>Substrate material</th>
<th>Cell density, cells/cm²</th>
<th>Open area, m²</th>
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(b) Graded cell reactor

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### TABLE II. DESCRIPTION OF FUELS

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<th>Naphthene</th>
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<th>Nitrogen</th>
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<td>Gross heat of combustion, kJ/kg</td>
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### TABLE III. RUN SEQUENCE

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<td>Naphthene</td>
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<td>4</td>
<td>ML-heavy blend</td>
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<td>3</td>
<td>Diesel (Run B)</td>
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<td>1</td>
<td>Naphthene</td>
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Figure 1. - Test section, hexagonal tube fuel injector shown.
(Dimensions in cm.)
Figure 2. - Fuel injector module. (Dimensions in cm.)

(a) HEXAGONAL TUBE MODULE (ONE OF 19).

(b) AIR-ASSIST MODULE (ONE OF 19).
Figure 3. - Air-assist fuel injector.
Figure 4. - Effect of fuel type on combustion efficiency.
Reference velocity, 10 m/s; pressure, 3x10^5 Pa.
Figure 5. - Effect of inlet temperature on combustion efficiency. Middle distillate fuel; reference velocity, 10 m/s; pressure, 3x10^5 Pa.

Figure 6. - Effect of reference velocity on combustion efficiency. Middle distillate fuel; inlet temperature 800 K; pressure, 3x10^5 Pa.
Figure 7. - Effect of fuel type on CO emission index.
Reference velocity, 10 m/s; pressure, 3x10^5 Pa.
Figure 8. - Effect of inlet temperature on CO emission index. Middle distillate fuel; reference velocity, 10 m/s; pressure, 3x10^5 Pa.

Figure 9. - Effect of reference velocity on CO emission index. Middle distillate fuel; inlet temperature, 800 K; pressure, 3x10^5 Pa.
Figure 10. - Effect of fuel type on unburned hydrocarbons emission index. Reference velocity, 10 m/s; pressure, $3 \times 10^5$ Pa.
Figure 13. \(\text{NO}_x\) emission index; reference velocity, 10 m/s; pressure, \(3 \times 10^5\) Pa.
Figure 14. - Effect of fuel type on NO$_x$ emissions.
Reference velocity, 10 m/s; pressure, 3x10$^5$ Pa.
Figure 15. Effect of reference velocity on NO\textsubscript{X} emissions. Middle distillate fuel; inlet temperature, 800 K; pressure, 3x10\textsuperscript{5} Pa.
Figure 16. - Effect of fuel on conversion of fuel nitrogen to NOx. Reference velocity, 10 m/s; pressure, 3x10^5 Pa.
Figure 17. - Effect of inlet temperature on fuel nitrogen conversion to NO₃. Middle distillate fuel; reference velocity, 10 m/s; pressure, 3x10⁵ Pa.
Figure 19. - Effect of reference velocity on pressure drop. Middle distillate fuel: inlet temperature, 800 K; pressure 3x10^5 Pa.
Figure 20. Fuel injector and reactor after mid-heavy blend tests.