A STUDY OF EMISSION INDICES WITH F76 AND FUEL ADDITIVES

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INTRODUCTION

On and off-road engine exhaust regulations have focused principally on new diesel and gasoline engines. There has not been much emphasis on regulating existing diesel engines or gas turbines used in mobile applications. However, off-road gas turbines and existing diesel engines may some day also be impacted by air pollution (NO_x, SO_x, CO, CO_2, particulates, UHC, etc) regulations.

Two problems arise when considering the possible regulation of existing and new marine engines in military use. The first is cost. Projected retrofit cost for emission controls on all of the Navy's existing engines would be high. The second deals with balancing power density against the emission of air pollutants. Navy marine applications, like most mobile applications thrive on high power densities (in both weight and volume). However, high power den-sities will typically be accompanied by high fir-ing temperatures. In turn, high firing tem-peratures are a precusor to high emissions of ther-mal NO_x. Thus the challenge is to improve power density without increasing NO_x generation to the environment.

The injection of ammonia (or related chemicals) in the exhaust stream of gas turbines or diesel engines is a very effective means for removing NO_x before it is released into the environment. This method can reduce NO_x emissions up to 90 percent in fossil-fueled power plants if the chemicals are applied within a critical temperature window. For mobile engines as those on ships, additional equipment and retention of chemicals for the processes are sometimes inconvenient, if not impractical.

One solution is to add the NO_x-removing chemicals directly to the fuel to avoid the need for carrying extra equipment or chemicals. The challenge with this approach is to protect the chemical additive as it passes through the combustion flame zone so that it can reduce NO_x in the exhaust stream. The Texaco Research Center in Beacon, New York has been working on such an approach for diesel engines. Texaco Inc. has produced some experimental fuels additives that have shown promise in reducing NO_x in diesel engines, but had not been evaluated in gas turbines.

The fuel (NATO F76) used by U.S. Navy ships must be suitable for both their diesels and gas turbine engines. This would also be a requirement for any fuel additives used by the Navy. Since previous fuel additive work has focused on diesel engines, a Navy program was initiated to explore the potential for these additives in gas turbines. The development of an additive suitable for both gas turbines and diesel engines was recognized as an unexplored research area, however if the Texaco fuel additive proves successful in all engines, then the challenge of power density vs NO_x generation would be met successfully.

PARTICIPANTS

with his assistant Doctoral candidate Li-keng Tseng.

ADDITIVE TECHNOLOGIES

The purpose of the fuel additive NO_x-reduction program was to assess whether certain fuel additive technologies that demonstrated NO_x reduction in compression ignition (diesel) engines could also reduce NO_x in turbine engine applications using F76 fuel combusted under simulated turbine conditions.

Three specific technology approaches were chosen:

1. Improve the burn characteristics of the fuel using a "combustion modifier".

2. Improve the dynamics of the fuel injector spray and diffusion using a "spray modifier".

3. Attempt to scavenge NO_x post-combustion using a "RAPRENOX-like" additive.

These technologies were chosen for the initial phase of this program since they demonstrated very favorable NO_x reduction results when evaluated in compression ignition tests using standard diesel fuels (Figure 1). The differences between the Navy's F76 fuel and the diesel used by Texaco were judged to be minimal. Thus, any change in NO_x formation between the diesel and gas turbine was postulated to result primarily from differences in the mode of combustion between the two engines and in differences in combustion temperatures.

F76 fuel was additized at the Beacon Research Labs of Texaco and NO_x emissions were evaluated at Purdue University in a simulated gas turbine combustor. Unadditized F76 fuel was used to generate baseline NO_x data; these data were compared with data generated from fuel containing the experimental additives. All additives evaluated at Purdue under simulated turbine conditions were first tested at Texaco under compression ignition conditions in die-sel fuel. Only those additives that demon-strated a net reduction in NO_x under compres-sion ignition were tested at Purdue. The addi-tives

investigated contain only carbon, hydrogen, oxygen, and nitrogen. Additives containing metals or metal salts were not in-cluded in the study because of possible en-vironmental hazards.

The specific samples of F76 fuel evaluated at Purdue are listed below:

Identification	Description
R96-1041.00	F76 baseline fuel
R96-1041.10	F76 fuel w/combustion modifier
R96-1041-20	F76 fuel w/combustion modifier
R96-1041.30	F76 fuel w/spray modifier
R96- 104 1.40	F76 fuel w/RAPRENOX-like additive

COMBUSTION MODIFIERS

Combustion modifiers are used to alter burn and time characteristics of the combustion process by chemically modifying the onset of ignition and the number of ignition kernels in the fuel spray.

In the first case, conventional combustion modifiers tend to advance the onset of diesel ignition, resulting in longer burn times and eliminating pressure spikes during the cycle. In turn, this decreases maximum in-cylinder com-bustion pressure and lowers maximum com-bustion temperature. Since NO_x generation is a direct function of combustion temperature, the lower temperature results in lower NO_x emissions in diesels. There appears to be no power loss associated with the use of com-bustion modifiers despite the change in both maximum pressure and rate of pressure increase compared to the base fuel.

In the second case, the modifier is thought to promote combustion not only in the premixed portion of the fuel spray, but also in the diffusion zone. Facilitation of combustion in the diffusion portion of the flame could also lead to a more controlled pressure increase and avoidance of pressure and temperature spikes.

Another explanation for the reduction of NO_x using combustion modifiers involves their interaction with the Zeldovich and/or the Prompt mechanisms of NO_x formation. Zeldo-vich and Prompt describe the formation of NO_x via the generation and reaction of radicals with

nitrogen, oxygen, and fuel under combustion conditions. Combustion modifiers are also thought to work via radical mechanisms. It can be postulated that the radicals formed by the modifier could interact with the Zeldovich and Prompt mechanisms in such a way as to suppress the formation of NO_x. The latter explanation is being investigated further for maximizing NO_x reduction.

Honda Engine Test Results
% NOx Reduction vs. Additive Thermal Stability
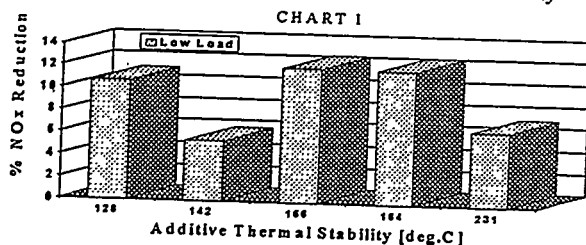
Honda Engine Test Results
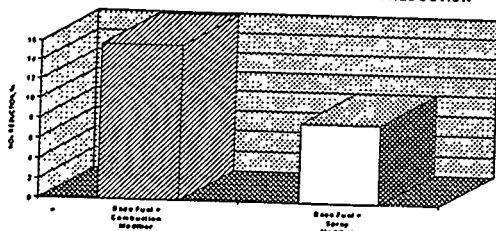### % NOx Reduction vs. Additive Thermal Stability
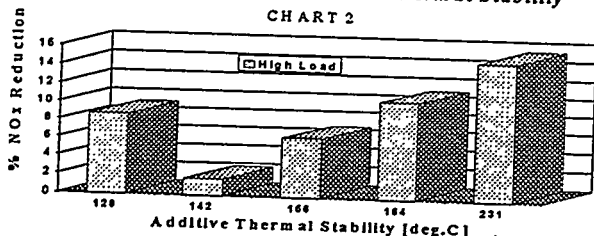
Figure 1: Effect of RAPRENOX-Like Additive (Top and Center) Combustion Modifier and Spray Modifier Additives on NO_x Reduction

The fuels referred to as R96-1041.10 and R96-1041.20 are F76 fuels that contain different combustion modifiers. In previous Texaco diesel engine tests, each additive decreased NO_x approximately 15 percent over that of the base fuel in diesel engine tests run at Texaco. These samples were submitted for NO_x testing under simulated turbine conditions at Purdue.

SPRAY MODIFIERS

Spray modifiers are used to alter physically the fuel injector spray patterns over those of the base fuel. In diesel engines these additives may have the same effect on combustion and emissions as increasing fuel injector pressure. Spray modifiers are expected to produce smaller droplet sizes (higher surface area to volume ratio) and increase swirl or turbulence of the fuel spray.

In diesel engine tests run at Texaco, spray modifiers have been shown to reduce NO_x emissions without increasing the cetane value of the fuel. Pressure/time curves, however, show similarities to cetane-improved fuels (Figure 2). In effect, spray modifiers advance the onset of ignition and can change the volume ratios of the premixed and diffusion portions of the fuel charge, resulting in longer burn times with lower pressure spikes during diesel combustion. A lower pressure spike produces lower maximum combustion temperatures and lower NO_x emissions. Typically, a 12 percent reduction in NO_x is observed in diesel combustion when comparing fuel containing a spray modifier with the base fuel (Figure 2).
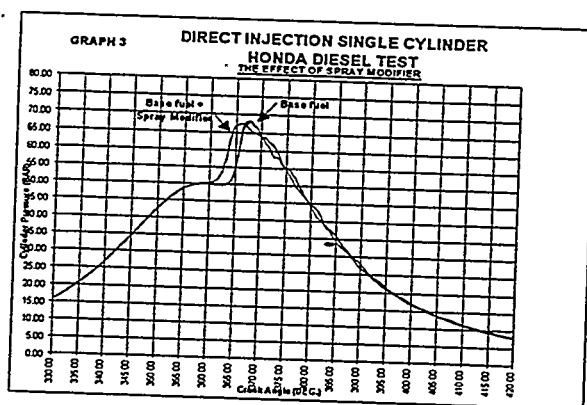

Figure 2: Effect of Spray Modifier on NO_x Reduction

Spray modifiers are temperature-sensitive, so it is necessary to match combustion temperatures with the modifier decomposition- temperature. Diesel combustion temperatures are generally lower than those in turbine engines,
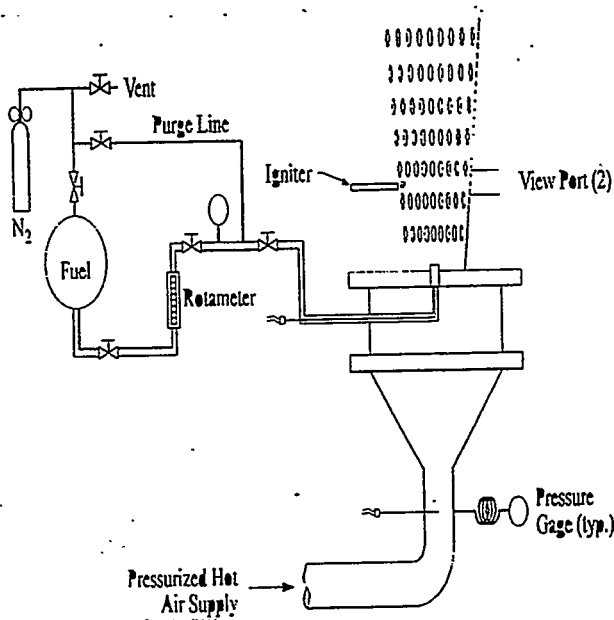
Figure 3: Experimental Apparatus

The facility used for the high-pressure tests is shown in Figure 4. A high-pressure air supply system was used as the preheated air source. Air pressure and temperature are monitored upstream of the combustor by a pressure gauge and a stainless steel, shielded, K-type thermocouple. The fuel supply system included a high pressure (up to 800-psi operating pressure) rotameter, which was calibrated for each tested fuel to ensure accuracy in fuel flow rate. The combustor was modified and reinforced to have an operating pressure up to 15 atmospheres, and a transparent plastic safe-to shield was added around the combustor section. A tail pipe with a diffuser plate was added to the system to provide a uniform subsonic exhaust stream from the system. Ten K-type thermocouples were welded to the outside surface of the combustor casing to monitor casing temperature during the high-pressure tests. The high mass-flow, high pressure air system could supply air at flow rates up to 9 lb/s at temperatures up to 750° F and pressures up to 2000 psi. The mass flow rate could be controlled with both subsonic and choked orifice plates, but for these tests a choked orifice plate was used, controlling mass flow by adjusting pressure upstream of the plate.
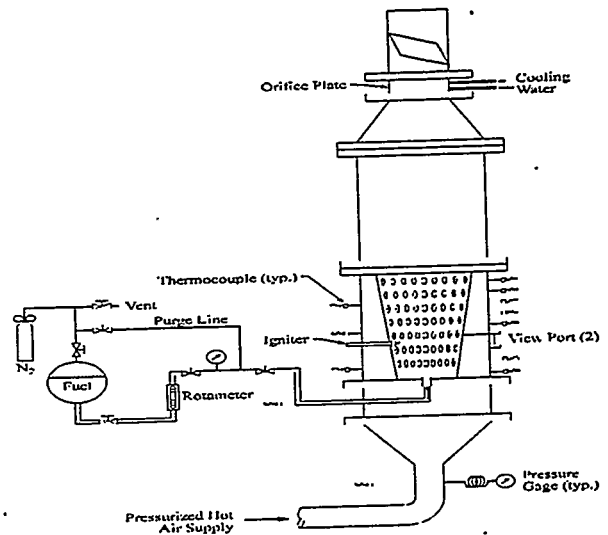

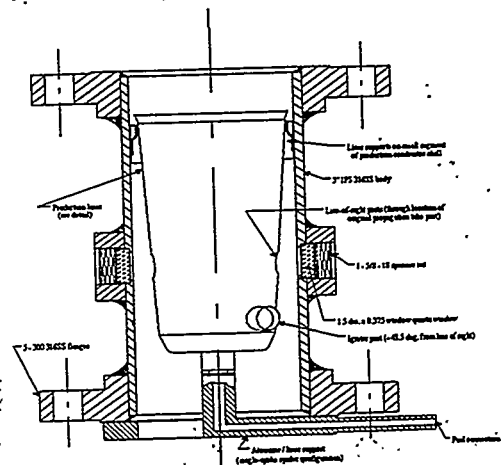**Figure 4: Schematic of High-Pressure Experimental Apparatus**

Figure 5: Schematic of Combustor

Figure 5 shows a detailed sketch of the combustor liner for a Boeing Gas Turbine Model 502-2E built for the Naval Bureau of Shipping (now the Naval Sea Systems Command). The diameter of the combustor is the same as the casing of the turbine engine so that a liner support segment from the original turbine engine could be fitted perfectly into the inside wall of the combustor. The fuel supply wafer is designed so that a minimum length of fuel line is exposed to the preheated air. A special igniter

was built to fit the igniter port. Two quartz windows are positioned at the exact location of the propagation port on the liner to provide visual access to the combustor. This design deviates slightly from the original combustor, but it provides necessary access for measurements and allows fuel emission properties to be examined under gas turbine conditions representative of Naval engines.

TEST PROTOCOL

The procedure for each test involved purging the fuel line after the appropriate fuel tank was connected to the fuel supply system. The air heaters were started after the air flow rate was adjusted to the desired test conditions. This heating process generally took about 2 hours since a large thermal mass was involved in the air supply system. During the heatup process, gas analyzers were warmed up and calibrated. After the preheated air was stabilized at the desired flow and temperature test conditions, the fuel supply system was pressurized and fuel injection and ignition in the combustor were initiated. The igniter was turned on before the fuel injection, and could be turned off after the fuel flow rate was adjusted to the desired value. The combustor was kept running at each test condition for about 15 minutes to reach steady-state conditions before global emission data were measured. Gas samples were taken continuously at the exit of the test apparatus. The sampled gases were delivered to NO_x, CO, and UIHC analyzers so that the concentrations of these pollutants could be measured simultaneously.

A schematic of the exhaust gas analysis system is shown in Figure 6. A water-cooled stainless-steel probe was used to draw samples from the exhaust into the gas analysis system. Concentrations of NO/NO_x were measured using a chemiluminescence-based analyzer (Thermo-Enviromental Instruments Model 10) with an accuracy as derived from the nitrous oxide (NO) or NO_2 calibration gas of 1 percent of full scale. Concentrations of CO and carbon di-oxide (CON were measured using a nondis-persiveinfrared (NDIR) analyzer

(Horiba MEXAGE 311) with a repeatability within 0.04 percent for CO measurements and within 0.3 percent for CO_2. The lowest (or least) count of the CO/CO_2 analyzer was 0.01 percent (100 ppm). This analyzer was calibrated using calibration gases of 5 percent CO_2, 2 percent CO, and 25 ppm NO. Dried nitrogen of 99.995-percent purity was used as the zero gas. A chiller-trap was used to condense water out before the sample entered the NO/NO_2 and CO/CO_2 analyzers. Since a CO analyzer with a lowest count of 0.01 percent is unsuitable for measuring of sub100 ppm CO levels, a gas-filter-correlation CO analyzer (Thermo-Enviromental Instruments Model 42) was used to measure CO concentrations in the 0- to 100-ppm range in some of the experiments. An HO analyzer, using a flame ionization detector (Rosemount Model 400A), was used to measure UHC concentration measurements in some of the experiments. The sampling line to the UHC analyzer was heated to prevent condensation.
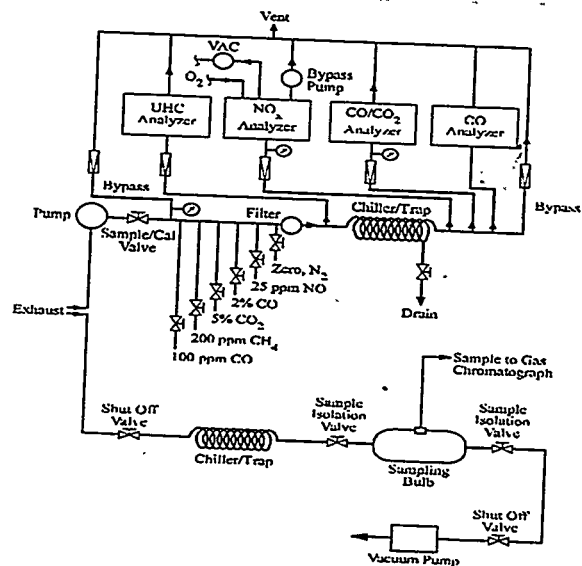

Figure 6: Schematic of Exhaust Gas Analysis System

TEST OBSERVATIONS

The test matrix included atmospheric pressure, 2-atmosphere pressure, and 15-atmosphere pressure tests. Tests at atmospheric pressure

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were first conducted for the experimental fuels to examine their atomization, vaporization, and combustion stability performance. The more promising experimental fuels were then tested at a combustor pressure of 2 atmospheres. Finally, one of the experimental fuels and the baseline fuel were tested at 15 atmospheres

Flame temperatures at atmospheric pressure for the baseline, R96-1041.30, and R96-1041.40 fuels were measured. Of these fuels, the add-tized R96-1041.40 had higher peak flame temperatures, particularly at higher equivalence ratios. Peak temperatures for the R961041.30 fuel were lower than those for the baseline fuel at lower equivalence ratios, but were similar at higher equivalence ratios

The R96-1041.10 fuel did not yield stable flames under most operating conditions. Under conditions that resulted in apparently stable flames, flameout would occur unexpectedly and erratically. Further testing of this additized fuel, therefore, was not pursued. The R96-1041.20 fuel yielded stable flames but caused significant precipitation of a solid powder in the fuel lines and on the fuel filter, eventually clogging the filter. Further examination revealed that this fuel also caused significant precipitation at room temperature in the fuel tank. The cause for the precipitation was not investigated, instead it was decided that tests with the R96-1041.30 fuel should be abandoned since precipitation of the additive could not be prevented. Stable flames and repeatable data were obtained for experimental fuels R96-1041.30, R96-1041.40 and the base-line fuel

The next phase of atmospheric pressure tests included fuels R96-1041.00, R96-1041.30, and R961041.40. Thermal stability tests were conducted for all three fuels up to 400°F. Fuel precipitation was not observed. This ensured that clean fuels were injected into the combustor since the fuel temperature before the fuel injector typically was about 175° F.

The additized fuel, R96-1041.30, when tested at 15 atmospheres reached stable condition, but a sudden change in emission readings and

combustor casing peaking temperature was observed. The peak temperature of the combustor casing increased to 580°C and, as a result, the test was terminated. During the combustor checkout after disassembling, it was discovered that the combustor liner had burned through in several locations during the high fuel-flow rate tests for R96-1041.30 fuel. After installing a new liner, high pressure tests were continued with a medium fuel-flow rate using the same test matrix and protocol as for the low flow rate tests. The flame still covered all the viewing area for both the baseline and the additized fuel. Emission data were obtained throughout the test matrix and the liner was not damaged when checked after post test disassembly.

A second series of tests was conducted at 15 atmospheres with the same test conditions used for the middle firing rate case. When R96-1041.30 was tested, it produced a much shorter flame than the baseline and the combustor wall temperatures rapidly increased near the dome of the combustor liner. A visual examination of the combustor liner showed slight damage to its inner surface. Surface roughness increased near the dome of the combustor liner, indi-cating that the flame was either impinging on or very close to the liner.

R96-1041.40 was tested next. The observations of the flame appearance and chamber temperature showed results similar to those of the previous high pressure tests of the same fuel under similar conditions, indicating that combustor performance had not been affected by the slight damage to the liner caused by the R96-1041.30 fuel.

RESULTS AND DISCUSSION

Two types of tests were conducted to cover the operating envelope of practical combusors. In the first type, the equivalence ratio was adjusted by changing the air flow rate while keeping the fuel flow rate constant. Under these conditions the equivalence ratio and the residence time varied simultaneously and significantly. The second type of tests involved varying the fuel flow rate to obtain

different equivalence ratios. Since residence time in the combustor is determined primarily by air flow rate, the results of these tests allowed the effects of equivalence ratio variations to be delineated

Figure 7 shows a comparison of Emission Index (EI) for the baseline fuel, R96-1041.00, and experimental fuel R96-1041.30 plotted as a function of equivalence ratio. The test conditions for Figure 7 were constant Fuel flow rate. Different fuel equivalence ratios were obtained by varying the air flow rate. This type of operation provides a wide range—at least a factor of three—of flow velocities inside the combustor. The characteristic residence time, therefore, also varies by more than a factor of three. It can be seen that the EINO and $EINO_x$ for R96-1041.30 are consistently higher than those of the baseline fuel, R96-1041.00. Both EINO and $EINO_x$ decreased as the fuel equivalence ratios increased for both experimental and baseline fuels. The reduction in available air reduces recirculation by changing the swirl number and causes jet-like luminous flames with higher equivalence ratios. The changes in flame structure lead to lower NO_x emissions with increasing equivalence ratio under these test conditions. The EINO and $EINO_x$ data for the two baseline fuel tests, one before testing experimental fuel R96-1041.30 and one after, are consistent. The differences in the EINO for the two tests of baseline R96-1041.00 fuel might have been caused by the fuel coking inside the combustor liner that was found after disassembling the combustor. This coking might have had a slight influence on CO and UHC measurements, but it would not have affected EINO and $EINO_x$ since the emissions they represent were produced in the flame zone.

To obtain comparable residence times, tests also were conducted with constant air flow rates, but fuel flow rates were varied to obtain a range of fuel equivalence ratios. One limitation of the tests with variable fuel flow rates is that three different nozzles had to be used to cover a reasonable range of equivalence ratios. Figures 8 and 9 show graphs of EI vs. fuel equivalence ratio for R961041.00
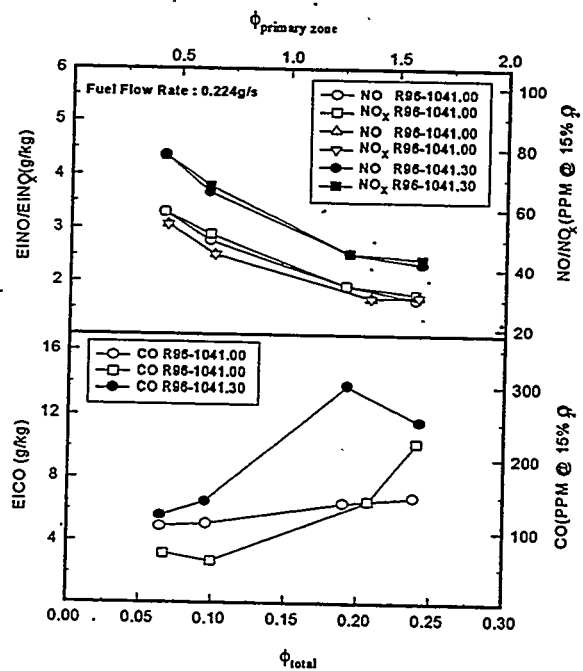

Figure 7: Pollutant Emissions for R96-1041.30 and Baseline Fuels at Constant Fuel Flow Rate Test Conditions Under Atmospheric Pressure

R96-1041.30, and R96-1041.40 feels under constant air flow rates. The EINO has the same values as the $EINO_x$ over these test conditions, so only the $EINO_x$ is shown. The flow field inside the combustor liner is relatively unchanged over the small range of fuel equivalence ratios for the constant air flow rate test conditions. The EI over this range of fuel equivalence ratios are relatively free from flow effects. Figure 8 shows that the $EINO_x$ slowly increases as the fuel equivalence ratio increases. This is because the product temperature also increases as fuel equivalence ratio increases. One can see that the $EINO_x$ remains about the same over each range except for the conditions at which larger droplets were created by the atomizers and EICO in-creased. As shown in Figure 9, the experimental fuel R96-1041.40 has higher $EINO_x$ at all test conditions.

In the first series of high-pressure tests, the baseline fuel and experimental fuel R96-1041.40 were tested with one air flow rate and three fuel flow rates at a combustor pressure of 15 atmospheres. The results show
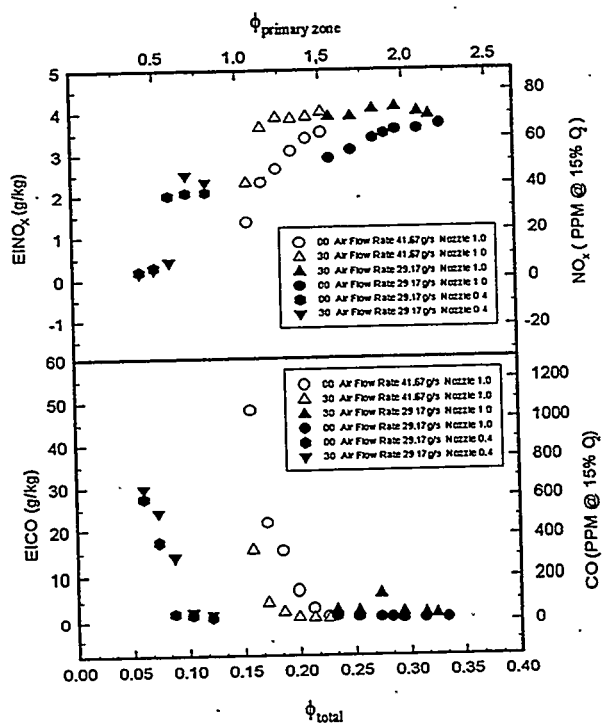
Figure 8: Pollutant Emissions for R96-1041.30 and Baseline Fuel at Constant Air Flow Rate Test Conditions Under Atmospheric Pressure

Figure 9: Pollutant Emissions for R96-1041.40 and Baseline Fuels at Constant Air Flow

that at these conditions R96-1041.40 still produces more NO_x than does the baseline fuel. Differences between the $EINO_x$ of the two fuels, however, decrease at this higher pressure, although the overall NO_x emissions increased for both fuels. This decrease in the difference between NO_x emissions for the baseline fuel and the experimental fuel may be due to a change in chemical reactions, change of combustor char-acteristics, or simply a decrease in the percentage of the extra NO_x produced by the fuel additives.

Figure 10 shows the EINO and $EINO_x$ as a function of fuel equivalence ratios for all 15-atmosphere tests, with open symbols representing data from the earlier tests and solid symbols representing data from the second series of tests. The baseline fuel F76 produces the lowest NO and NO_x emissions for all test conditions. The difference between NO_x emissions of the R961041.40 fuel and the baseline fuel is similar for the earlier and present tests
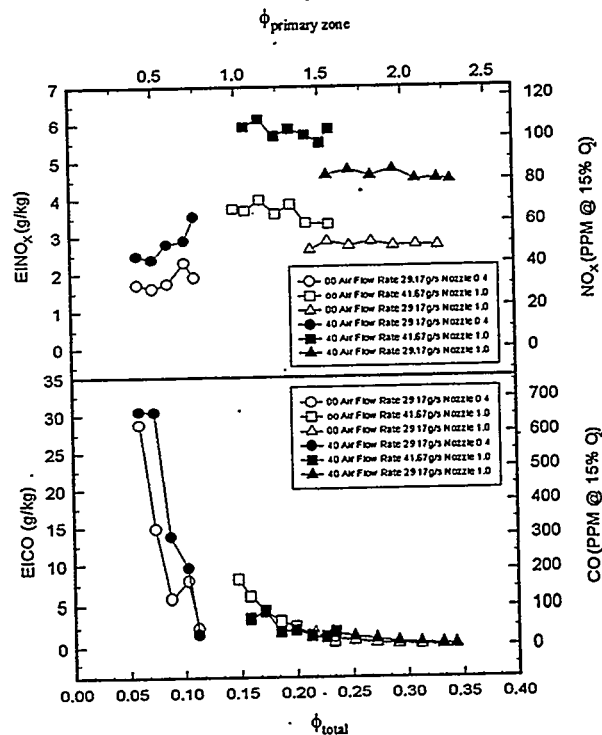
except for the data obtained at the high firing rate, high fuel-equivalence ratio, where flame impinging on the combustor liner could be the cause of the small difference. The flame impingement also caused a burn-down of a combustor liner during the earlier tests. The fuel mixture, an equal volume mixture of R96-1041.30 and R96- 1041.40 fuels, shows EINO and El NO_x similar to those of the R96-1041.40 fuel. The EINO and $EINO_x$ produced by R96-1041.30 fuel have a value between the baseline F76 fuel and the R96-1041.40 fuel. The R96-1041.30 fuel, however, also caused a dramatic change of temperature distribution in the combustor liner, which resulted in minor damage to it.

The effects of the R96-041-30 (spray-modifier) fuel additive (microexplosive) were further examined in a spray flame in a co-flow air configuration. A pressure swirl atomizer with a flow rate of 0.4 gallon per hour was used as the fuel injector for the spray flames. The fuel injector was placed in an axisymmetric co-flow of air with a duct diameter of 3 inches. The
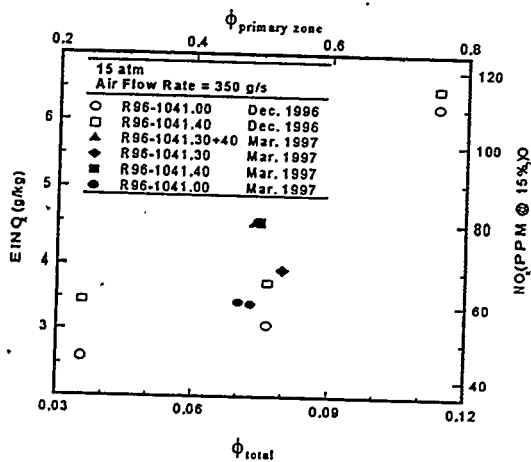
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Figure 10: NO$_x$ Emissions at 15 Atmospheres at 3 meters per second.

velocity of the co-flow air was kept constant.

The fuel supply pressure was also kept constant at 110 psig during the test. Two different fuels, baseline F76 (R96-1041.00)) fuel and the R96-10)41.30) fuel, were tested under identical test conditions.

Figure 11 shows photographs of the two spray flames under identical operating conditions. The spray flame shown on the bottom is fueled by baseline F76 fuel and the one on the right is fueled with the R96-1041.30 fuel. One can see that the F76-fueled spray name has a flame height about twice as long as the spray flame fueled with the R96-1041.30 fuel. This is be-cause the F76-fueled spray name has a larger fuel droplet size and. therefore. longer evapora-tion time is needed to vaporize these larger fuel droplets. The spray-modifier fuel additive in the R96-1041.30 fuel causes the larger spray droplets produced by the pressure atomizer to experience a microexplosion and reduces them to smaller droplets. The lifetime of the smaller droplets is much shorter and so the height of the spray flame fueled by the R96-1041.30 fuel is much shorter. The angle of the F76-fueled spray flame is smaller than that of the R96-1041.30-fueled spray flame.

Two possible mechanisms contribute to this phenomenon:

1. The microexplosion caused by the spray modifier additives in the R96-1041.30 fuel increases the spray angle

2. The momentum transfer for the smaller droplets in the R96-1041.30 fuel spray flame is much more effective than that for the larger droplets in the F76-fueled spray flame.

Although the spray-modifier led to an increase in NO$_x$ in the present combustor configuration. advanced combustor concepts such as lean direct injection (LDI) would benefit significantly from the use of this additive.
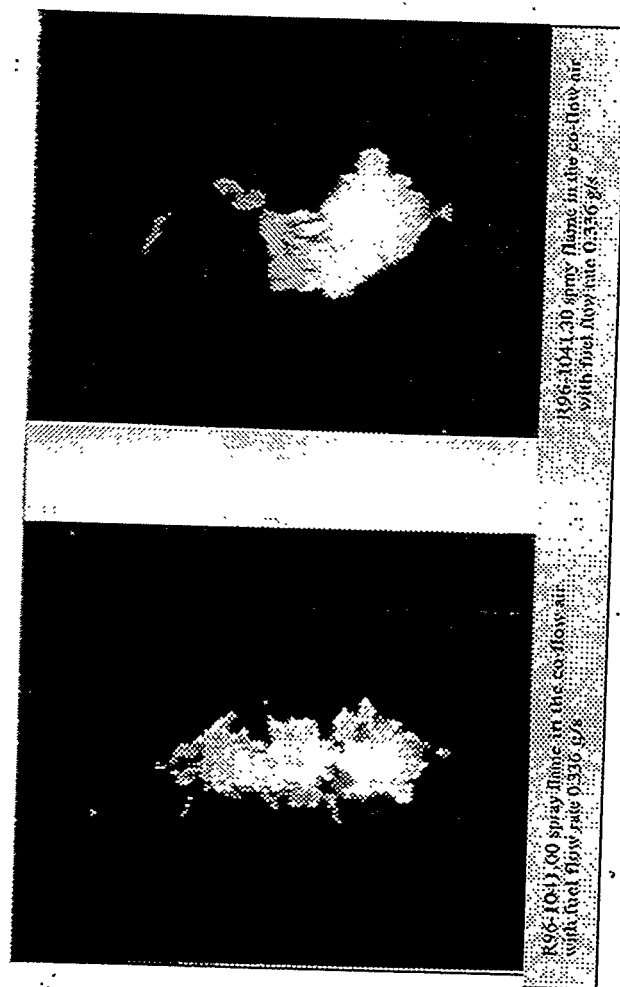

Figure 11: Spray Flames

CHEMICAL REACTION MODEL

The effects of adding the nitrous oxide-(NO$_x$-) reducing chemicals were studied computationally using an opposed-flow non-premixed flat-

flame code with methane as the fuel. This one-dimensional flame provides an opportunity to conduct a detailed study of the chemical reac-tion mechanism within the current computation capacity. The most recent reaction mechanism developed by the Gas Research Institute (GRIMECH) was used for this study. The mechanism considers 49 chemical species and 279 reversible reactions.

Figure 12: QRPD for Methane/Air Flames With Initial Temperature = 600° K

Figure 12. shows the QRPD of important NO_x emissions-related reactions for the nonad-ditized and the HNCO-added flames with an initial temperature 600° K. Each line in the QRPD represents a chemical reaction. The line thickness representing the value of the integrated reaction rate of this specific reaction over the entire flow field. The direction of reaction is indicated by the arrows. The reaction is producing or consuming a species if the arrow is pointing into or away from the species

respectively. It can be seen that the additive HNCO is mainly involved in the reactions with HCNO, NO, and NH_2. A comparison of the upper panel and the lower panel of Figure 12 shows that the reaction rate of many NO-related chemical reactions are drastically aug-mented. Reaction number 265 was plotted in one tenth of the scale to fit into the plot. Adding HNCO not only increases the con-sumption rate of NO but also increases the production rate of other species such as NH, HNO, and nitrogen (which all lead to more NO), and so results in a net increase of NO produc-tion.

CONCLUSIONS

Measurements of pollutant EI at three different pressures for baseline F76 and four additized fuels were attempted. Of the four additized fuels, R96-1041.10 did not yield stable flames under gas turbine conditions and R96-1041.20 fuel showed solid precipitation at room tem-perature in the fuel line and in the fuel filter. The reasons for these unexpected results were not pursued, thus it is possible that the addi-tives may have been incorrectly formulated.

R96-1041.30 and R96-1041.40 fuels showed approximately a 30- to 50-percent increase in NO_x emissions over the baseline fuel index at combustion pressures of 1 and 2 atmospheres. At 15 atmospheres pressure, the $EINO_x$ for R96-1041.40 fuel is 30 percent higher than the $EINO_x$ baseline fuel under low and medium fire conditions. The $EINO_x$ for the two fuels are almost identical for the high fire conditions, but flame impingement on the liner may have af-fected these data.

The results from present tests suggest that the fuel additives may become more effective at pressures higher than 15 atmospheres, but this is conjecture and needs to be verified directly. Therefore, tests in the pressure range of 18 to 30 atmospheres should be explored.

Despite the higher NO_x emissions with the additized fuels, the fact that the emissions were affected would indicate that tailoring of the additive for a gas turbine combustor may
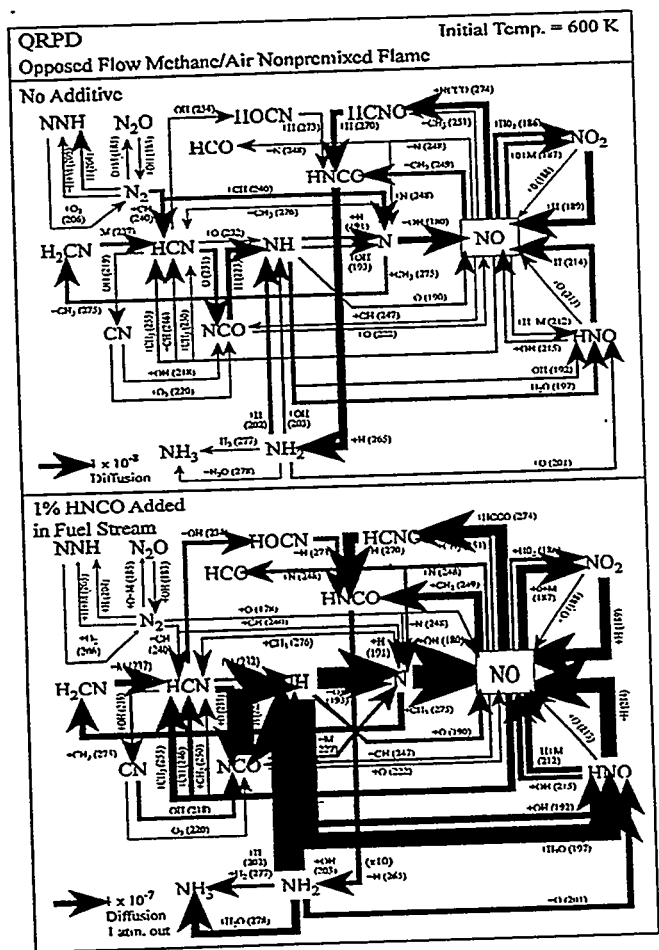
be more successful.

The fuel additives appear to affect the atomization performance, so the surface tension and viscosity changes that they may cause need to be examined. The atomization characteristics need direct experimental investigation.

The reductions in NO_x emissions caused by the fuel additives were in the 10- to 20-percent range based on diesel engine tests at Texaco Inc. This was the first time these additives were evaluated in a gas turbine combustor. The results of this evaluation point out the complexity in using fuel additives in two different combustion processes.

Lean Direct Injection (LDI) combustor concepts may find value in the additives similar to R96-1041-30 since this additive seems to promote a more rapid combustion.