Pollutant Formation in Heterogeneous Mixtures of Fuel Drops and Air

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The aim of this study is to determine changes in emission characteristics, particularly soot and NO_x formation, arising from changes in fuel spray characteristics. Tests are conducted in a tubular air casing containing a 15 cm diam combustor liner. Diesel oil (DF 2) is supplied through a circular array of 68 airblast atomizers mounted within the test section on a sliding rod, which permits axial movement of the fuel injectors relative to the plane of gas sampling. Mean fuel-drop size is varied by adjusting the air flow rate to the airblast atomizers. The flame is stabilized on a perforated-plate flameholder. Samples are collected through a stainless steel probe equipped with water cooling and nitrogen injection. Equivalence ratio is varied from 0.5–1.3, while maintaining the inlet air pressure and temperature constant at 507 kPa (5 atm) and 573 K, respectively. Results are presented to demonstrate the influence of mean fuel drop size, flame length, and equivalence ratio on pollutant emissions.

Introduction

IN order to exploit fully the potential advantages of stratified charge combustion in terms of improved fuel economy for both spark ignition and diesel engines, more knowledge is needed on the influence of fuel type, mean drop size, and vapor concentration on burning rates and on the formation of various pollutant emissions, such as particulates, carbon monoxide, unburned hydrocarbons, and oxides of nitrogen. More knowledge in this area would promote optimal engine design and provide useful data for the validation of mathematical models of flame propagation and emissions formation in both reciprocating and gas turbine engines. Further incentive is provided by the need for a better fundamental understanding of the basic role of fuel chemistry in determining the amount and nature of the particulate emissions from internal combustion engines. Such knowledge is especially important for the alternative fuels that are now being actively investigated. Unfortunately, when the combustion characteristics of alternative fuels are examined in an actual engine, the effects of the different physical properties cannot be distinguished from the effects arising from the different chemical properties. Thus, the results obtained on any given engine cannot be readily extrapolated to other engines or other operating conditions.

The present research has the following main objectives: 1) By precise control of fuel drop size and drop size distribution, the effects of changes in physical properties are eliminated, so that any changes in combustion characteristics (e.g., burning rates, particulate formation, etc.) arising from a change in fuel type can be attributed directly to chemical effects; 2) To provide basic data on important combustion properties such as the optimum fuel drop size for minimum particulate formation.

Experiment

The basic test facility is shown in Fig. 1. The working section consists of a tubular air casing containing a tubular combustion liner 15 cm in diameter. A unique feature of the test rig is the mode of fuel injection. This comprises an array of 68 "microscopic" airblast atomizers, which are mounted on a sliding rod to permit axial movement of the fuel injectors relative to the plane of sampling of emissions. Also mounted on the same rod and in the same plane as the atomizers is a flat plate containing a number of holes and slots to allow passage of the fuel drops and combustion air, as shown in Fig. 2. This flat-plate flameholder is made of Hastelloy X. The location of each atomizer within the liner was determined by dividing the liner into sections of equal area and locating a fuel injection point as close as possible to the center of each section. The objective of this arrangement is to achieve a sensibly uniform mixture strength in the fuel-air mixture entering the flame zone. This, in turn, ensures uniform conditions of temperature and chemical composition within the combustion zone at any given axial distance downstream of the fuel injection plane.

A further important advantage of this fuel injection system is that it allows the mean drop size of the fuel spray to be varied in a controlled manner, while maintaining all other flow conditions constant. Thus, the influence of fuel drop size on the formation of soot and other pollutant emissions may be readily determined. Furthermore, when carrying out tests on the effects of fuel composition on exhaust emissions, it is possible to adjust the atomizing airflow rate so as to achieve a constant drop size for all fuels. Alternatively, based on a knowledge of the density, viscosity, surface tension, and evaporation constant for each fuel, it is possible to select a mean drop size that will ensure a constant rate of evaporation for all fuels. This means that all observed differences in exhaust emissions can be attributed directly to variations in chemical properties, such as aromatics content or hydrogen content. Thus the method of fuel injection constitutes a powerful tool for separating the effects of physical properties and chemical properties when assessing the propensity of any given fuel toward soot and particulate formation.

The fuel employed in this investigation is a light diesel oil, DF-2. The main properties of this fuel are the following: density = 860 kg/m^3, viscosity = 0.0026 kg/(ms), surface tension = 0.027 kg/s^2, and boiling range = 444–630K.

Received April 22, 1986; presented as Paper 86-1526 at the AIAA/SAE/ASME/ASEE 22nd Joint Propulsion Conference, Huntsville, AL, June 16–18, 1986; revision received July 23, 1986.

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Measurement of Emissions

The exhaust sampling probe, as illustrated in Fig. 3, is located at the combustor axis. It is constructed of stainless steel, is water-cooled, and features static pressure and nitrogen injection ports near the tip. The probe tip is also water-cooled, but is made of a special temperature-resistant alloy, similar to Hastelloy.

The probe cooling water, supplied at high pressures (0.7–2.8 MPa), flows in the annulus created between the inner (0.40 cm diam) sample tube and the concentric middle (0.79 cm diam) tube. The clearance between the middle tube and the probe tip is made small (0.05 cm) to insure that the probe tip is properly cooled. Mellor and Clark[1] found that measured unburned hydrocarbons (UHC) and CO concentrations can be affected by the presence of a tapered, sharp probe tip due to inadequate cooling, which prevents immediate quenching of reactions within the probe. The cooling water is discharged through the annulus between the middle tube and outer wall of the probe.

The probe dimensions represent a compromise between the need for small tip geometries to minimize flow disturbances, and the requirement for adequate probe inlet area to facilitate soot particle entry. The internal diameter of the probe is relatively large (0.3 cm) to alleviate anticipated clogging problems. Its frontal cross-sectional area is 0.5% of that of the combustor, following the recommendation of Bilger.[2] The probe tip geometry used here is based on research by Rouillard and Hicks,[3] where they showed that dust probes with internally beveled tips and constant outside diameters greatly minimized upstream disturbances.

No attempt was made to sample isokinetically in this study. It has been argued that isokinetic sampling is required to produce representative soot samples,[4] but numerous studies on gas turbine and diesel particulates have shown that sampling rate has a negligible effect on particulate concentrations.[5-8] A brief check on the effect of sampling rate supported this conclusion.

Kadota and Henein,[9] and more recently Clark et al.,[5] have shown that the addition of nitrogen to the sample gas increases the particulate yield by preventing water condensation. Therefore, a nitrogen injection port was included in the probe tip to further investigate this effect. However, all the particulate data presented here were obtained without nitrogen injection.

Gas Analysis System

The exhaust gases are continuously analyzed for concentrations of carbon monoxide (CO), carbon dioxide (CO_2), unburned hydrocarbons (UHC), nitric oxide (NO), nitrogen dioxide (NO_2), and oxygen (O_2) using infrared analyzers, a flame ionization detector, a chemiluminescent analyzer, and a paramagnetic analyzer, respectively. Exhaust gases are drawn from the combustor into the gas analysis system through electrically heated sampling lines. The sampling lines consist of a teflon inner tube, which is surrounded by a stainless steel mesh covered by insulation, and a resistive heating element. The temperature of the 6 m sampling line is maintained at 150°C to prevent water condensation. Two heated filters remove particulate matter from the system, and a cold trap prevents water vapor from reaching the infrared, paramagnetic, and chemiluminescent analyzers. The flame ionization detector is equipped with an internal filter.

A dilution tunnel is used in the measurement of particulate concentrations. It produces a representative particulate sample by mixing the combustion products with an excess of heated dry air, supplied at a constant flow rate. Particulate samples are then collected downstream of the dilution region on a filter paper. Descriptions of dilution tunnel theory and operation can be found in publications by Gillette,[10] Dunleep,[11] Frisch et al.,[12] and MacDonald et al.[13]

Particulate Sampling Procedure

Combustion is initiated at atmospheric pressure. Fuel and air flow rates are then slowly increased as the combustor pressure rises. When the desired operating pressure has been reached, adjustments are made to obtain the correct inlet air temperature, velocity, and equivalence ratio. During this period, the dilution tunnel pressure is maintained well above the test section pressure to "back flush" the system and prevent probe clogging and sampling line deposition. Next, a filter paper is placed into the holder, and the dilution tunnel pressure is lowered by opening the back pressure valve. Finally, the appropriate dilution air flow rate is established.

Each particulate data point shown is actually an average of three separate measurements. Once the dilution flow rate is established, the pneumatically actuated ball valve is opened and the particulate sample flows through the filter paper and flow meter apparatus. The sampling period usually lasts about twenty seconds during which time 1–4 mg of particulates are deposited, depending on the conditions. The ball valve is then closed and the volume of sample gas is recorded along with the CO_2 concentration of the mixture. This procedure is immediately repeated for two additional filters.

The filters are dried in a temperature and humidity controlled environment for eight hours before and after the sampling run. Their weights are measured to determine the amount of particulate deposited during the sampling period.

Mean Drop Size

Before starting on the main combustion research program, a series of tests was conducted to examine the influence of combustor operating conditions on the mean drop sizes produced by the array of airblast atomizers. Mean drop size and drop size distribution were measured across the centerline of the spray under cold flow conditions, using a Malvern particle

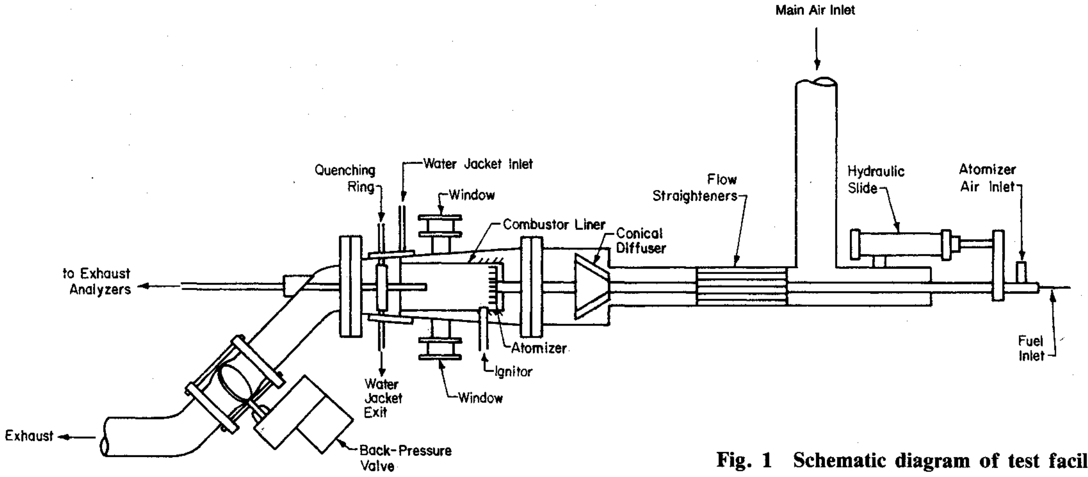

Fig. 1 Schematic diagram of test facility.

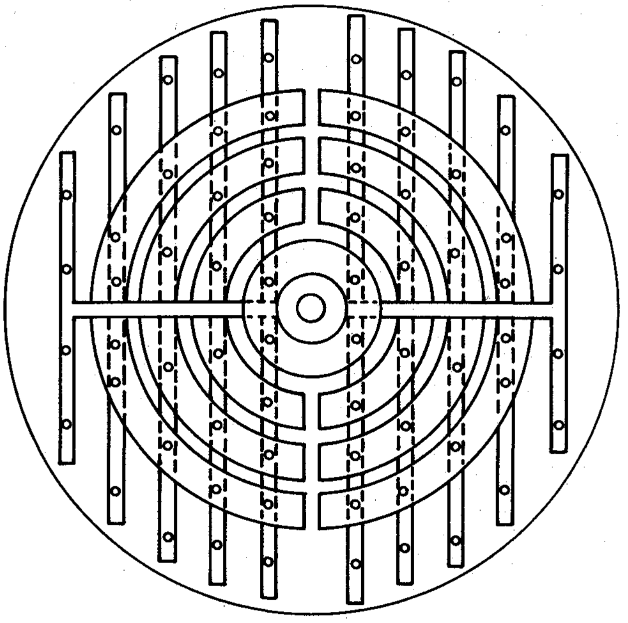

Fig. 2 Probe for emissions sampling.

sizer. The results of this study have been published elsewhere.[14] For a wide range of conditions, it was found that mean drop sizes (SMD) could be correlated with good accuracy by the equation

$$SMD = 0.00365\left[\frac{\rho_L \sigma}{\rho_A^{0.5} U_R^2}\right]^{0.5}\left[1+\frac{1}{ALR}\right]^{0.7}$$

where ALR = air/liquid mass ratio; U_R = relative velocity between atomizing air and liquid, m/s; ρ_A = air density, kg/m^3; ρ_L = liquid density, kg solidus/m^3; and σ = surface tension, kg/s^2.

Results

All the experimental work was conducted at a pressure of 507 kPa (5 atm), and an inlet air temperature of 573 K. These levels of pressure and temperature are high enough to ensure that burning rates are not limited by chemical reaction rates, and that the level of combustion efficiency is always close to 100%, regardless of fuel drop size. At the same time, the thermal loading on the liner walls is not high enough to cause any appreciable problems of wall buckling and overheating.

Some of the results obtained at these conditions are shown in Figs. 4–12. Figure 4 illustrates the variation of oxygen concentration with equivalence ratio at distances downstream of the flameholder of 13, 17, 21, and 25 cm. In effect, Fig. 4 provides an indication of the rate of combustion. The carbon dioxide concentration also gives a useful indication of the rate of chemical reaction, and Fig. 5 serves to demonstrate that, for near-stoichiometric mixtures, combustion is complete within a few inches downstream of the flameholder. Figure 6 illustrates the variation in UHC concentrations with equivalence ratio for three values of mean drop size. All three curves have the same general characteristics. They show that UHC emission diminish with increase in equivalence ratio up to around the stoichiometric value, beyond which any further increase in equivalence ratio causes UHC emission levels to rise to very high values. These trends are typical of those obtained with other types of continuous combustion systems.[15] The high levels of UHC at low equivalence ratios represent incomplete combustion of the fuel, due to the low reaction temperatures. At higher equivalence ratios ($\phi > 1.0$), large amounts of UHC are formed owing to the lack of sufficient oxygen to complete the reaction to CO_2 and H_2O. Figure 6 also serves to show that, at equivalence ratios greater than 0.6, improvements in atomization quality produce appreciable reductions in UHC. At equivalence ratios less than 0.6, variations in mean drop size appear to have little effect on UHC emissions. This is because increase in evaporation rates, arising from reduction in mean drop size, offers no advantage at low equivalence ratios where rates of combustion are limited, not by evaporation rates, but by chemical reaction rates.

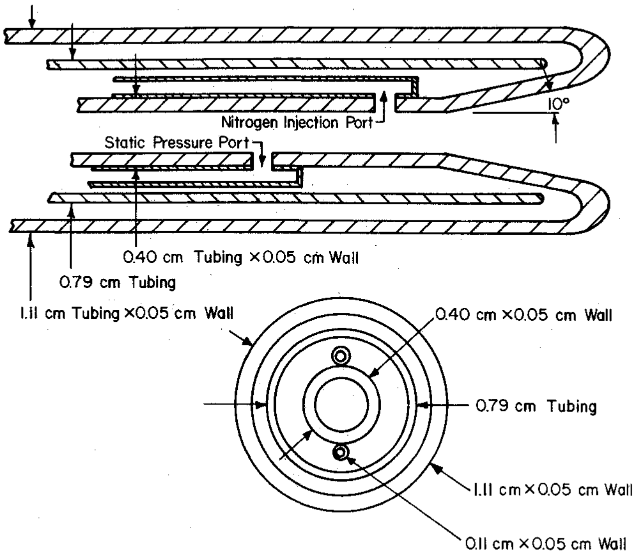

Fig. 3 Influence of flame length and equivalence ratio on oxygen concentration in exhaust gas.

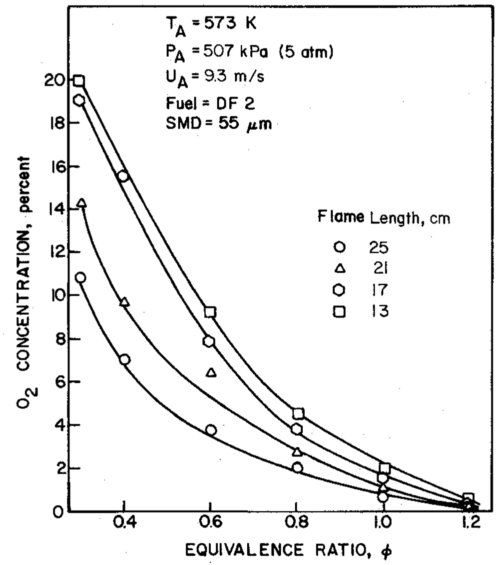

Fig. 4 Influence of flame length and equivalence ratio on carbon dioxide concentration in exhaust gas.

As the fuel-bound nitrogen content of the DF-2 employed in this study is negligibly small, the nitric oxide found in the exhaust gas is produced mainly by the oxidation of atmospheric nitrogen in high-temperature regions of the flame. The process is endothermic, and it proceeds at a significant rate only at temperatures above about 1800 K. Generally, it is found that NO concentrations peak on the fuel-lean side of stoichiometric, and this is clearly observed in Figs. 7–10. Figures 7 and 8 both show that increase in flame length lead to higher NO emissions. This is due partly to the increase in flame temperature with increase in downstream distance from the flameholder, and also to the longer residence time created by increase in flame length. As the production of nitric oxides is

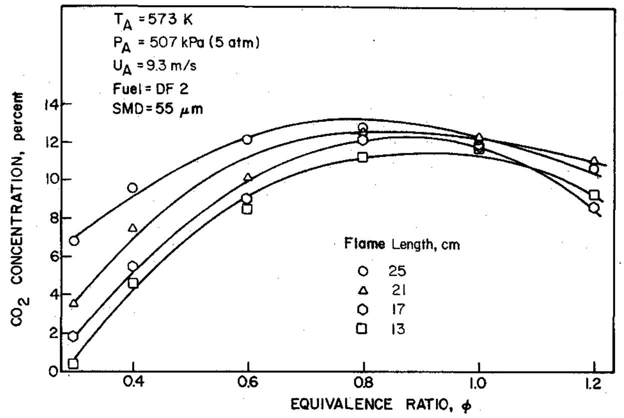

Fig. 5 Influence of mean drop size on unburned hydrocarbon emissions.

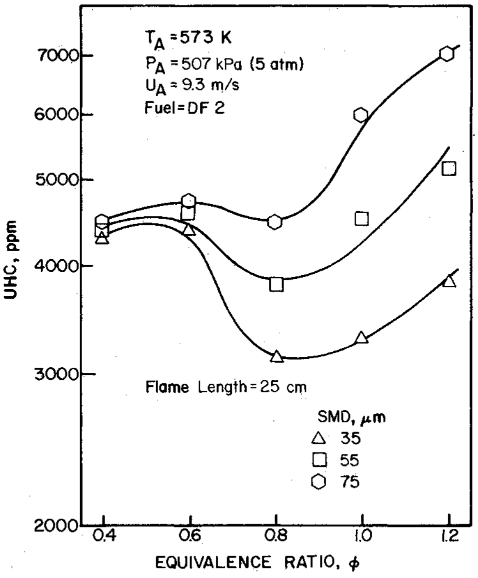

Fig. 6 Influence of flame length on NO emissions (SMD = 35 μm).

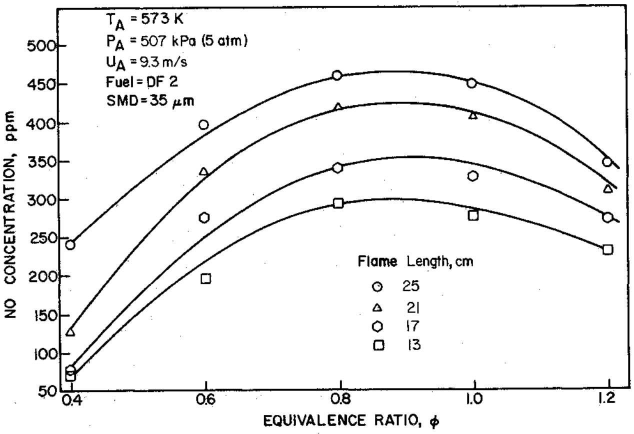

Fig. 7 Influence of flame length on NO emissions (SMD = 55 μm).

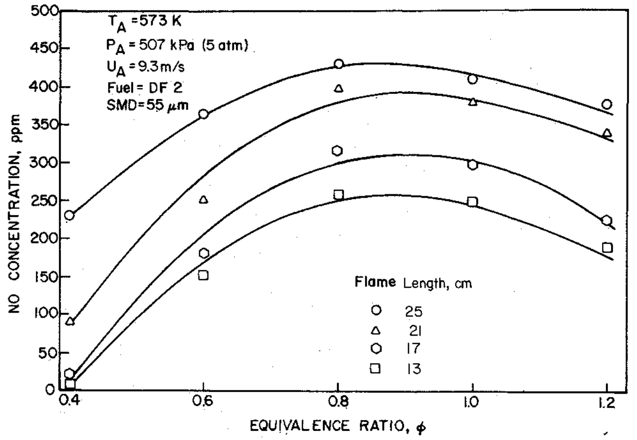

Fig. 8 Influence of mean drop size on NO emissions (flame length = 17 cm).

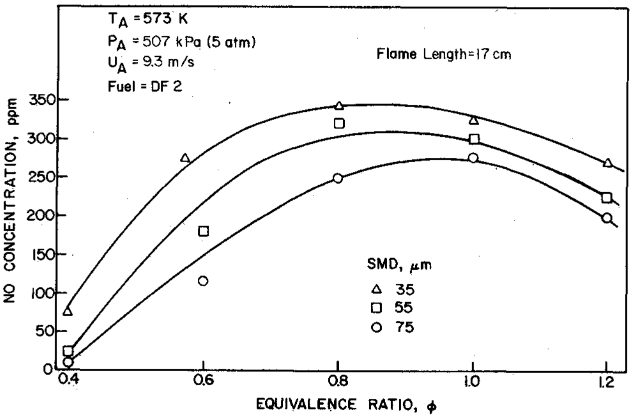

Fig. 9 Influence of mean drop size on NO emissions (flame length = 21 cm).

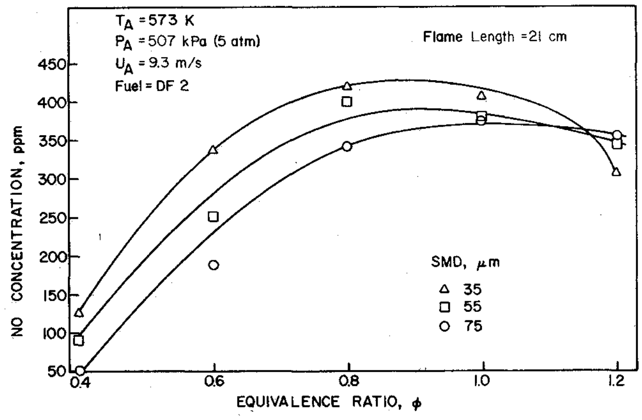

Fig. 10 Influence of flame length and equivalence ratio on particulate emissions (SMD = 55 μm).

exponentially dependent on temperature and linearly dependent on time, the effects of higher temperature and longer residence time combine to raise the measured values of NO. The differences in the measured levels of NO between Figs. 7 and 8 are due solely to differences in mean drop size, the SMD being 35μm for the data in Fig. 7 and 55μm for the data in Fig. 8 The influence of atomization quality on NO emissions is shown more directly in Figs. 9 and 10. These figures demonstrate that reduction in mean drop size causes NO emissions to increase. This is because small drops evaporate faster than large drops so that fuel evaporation is complete closer to the flameholder. For a fixed overall length, this means that combustion starts earlier and the combustion products spend more time at high temperature. At the very high pressures and temperatures encountered in modern gas turbines and diesel engines, evaporation times are much shorter than for the less arduous conditions of the present experiments. In consequence, the influence of mean drop size on NO emissions is appreciably less.[15]

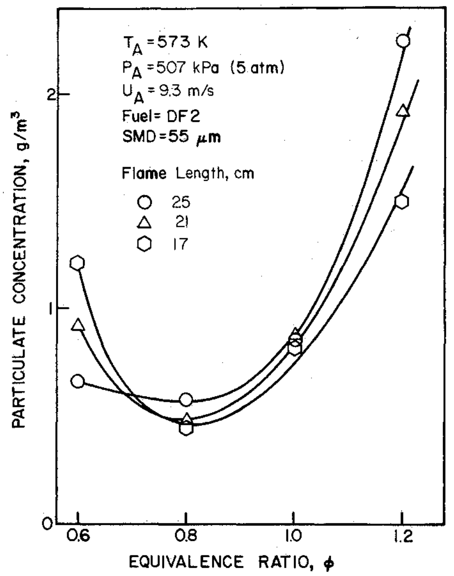

Fig. 11 Influence of flame length and equivalence ratio on particulate emissions (SMD = 55 μm).

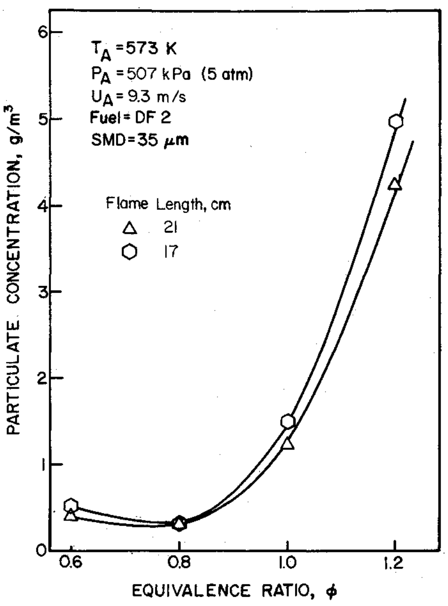

Fig. 12 Influence of flame length and equivalence ratio on particulate emissions (SMD = 35 μm).

Particulate emissions are caused mainly by the production of finely divided soot particles in fuel-rich regions of the flame. From a soot viewpoint, a combustor may be considered to comprise two separate zones, a soot formation zone and a soot consumption zone. The soot concentration actually observed in the exhaust gases represents the dominance of one zone over the other. This two stage nature of soot production is clearly illustrated in Fig. 11. At low equivalence ratios rates of soot formation is slow and, in consequence, very little soot is formed. However, temperatures are also very low, so that most of the soot formed is retained in the exhaust gases. Increase in equivalence ratio toward unity leads to higher flame temperatures and higher reaction rates, so that the rate of soot consumption increases and the soot concentration in the exhaust gas goes down. With further increase in equivalence ratio beyond unity, the rate of soot formation rises very rapidly due to insufficient oxygen to oxidize the fuel to carbon monoxide and hydrogen.

Figure 12 exhibits the same general trends as Fig. 11 but, for equivalence ratios less than 0.8, the levels of particulate concentration are lower. As the only difference in test conditions between Figs. 11 and 12 is the mean drop size of the spray, the lower particulate concentrations shown in Fig. 12 must be due to the finer spray 35μm as opposed to 55μm in Fig. 11. The importance of mean drop size to particulate formation is due to the fact that, as the fuel spray approaches the flame front, heat transmitted from the flame starts to evaporate the drops. The smaller drops in the spray have enough time to evaporate completely ahead of the flame zone, and the fuel vapor generated thereby mixes with the combustion air and burns as a premixed flame. However, the larger drops in the spray do not have sufficient time to evaporate completely and they burn as individual drops, each of which is surrounded by a diffusion flame. With a premixed flame, soot formation does not occur until the amount of available air is reduced to about 65% of the stoichiometric value. However, for the diffusion flame surrounding a droplet, soot formation is dependent only on the fuel and the size of the drop. Thus, under these conditions, even where ample oxygen is available, particulate emissions are dependent on the initial mean drop size of the spray.

Comparison of Figs. 11 and 12 shows that, for fuel-rich mixtures, improvements in atomization quality cause particulate emissions to increase. This is because, in the combustion of fuel drops, the fuel/air ratio that governs chemical reaction rates and soot formation is not the "overall" fuel/air ratio, which is obtained as the total mass of fuel divided by the mass of air, but the "effective" fuel/air ratio, which is given by the ratio of the mass of *fuel vapor* to the mass of air. For any given overall fuel/air ratio, a reduction in mean drop size will promote more rapid evaporation and thereby increase the effective fuel/air ratio in the combustion zone. Thus, in comparing Figs. 11 and 12, the particulate concentration in Fig. 12 is found to be higher at equivalence ratios in excess of unity. This is due to the higher effective fuel/air ratio in this region, stemming from the more rapid evaporation of the smaller fuel drops.

Conclusions

Improvements in atomization quality lead to worthwhile reductions in the emissions of unburned hydrocarbons at equivalence ratios higher than around 0.6. At lower equivalence ratios, where rates of combustion are limited mainly by chemical reaction rates, the enhancement of evaporation rates resulting from a reduction in mean drop size, has little effect on the combustion process, and unburned hydrocarbon emissions are sensibly independent of mean drop size.

The measurements of oxides of nitrogen generally confirm previous conclusions in showing that increase in flame length leads to higher exhaust gas concentrations of NO. Reduction in mean drop size causes NO emissions to increase, and this is attributed to the corresponding reduction in the time required to evaporate the drops, which allows the NO reactions more time to proceed toward their equilibrium concentration.

Reduction of mean drop size in the fuel spray leads to lower emissions of particulates for fuel-weak mixtures, and to higher particulate emissions for fuel-rich mixtures. This is because, for weak mixture strengths, reduction in mean drop size enhances evaporation rates which causes a larger proportion of the fuel to be burned in the mode of a premixed flame. However, for fuel-rich mixtures, the higher rates of fuel evaporation associated with smaller drops, lead to higher rates of soot formation by increasing the fuel vapor/air concentration in the combustion zone.

Acknowledgment

The support of this work by the Department of Energy, with Dr. Ron Graves and Dr. Ralph Fleming serving as technical monitors for the project, is gratefully acknowledged.

References

[1] Mellor, A. M. and Clark, J. A., "Probe Effects in Gas Turbine Combustion Emission Measurements," ASME Paper 80-GT-71, 1980.

[2] Bilger, R. W., "Probe Measurements in Turbulent Combustion," *Combustion Measurements,* edited by R. Goulard, Hemisphere Press, Washington, D. C., 1976.

[3] Rouillard, E. E. A. and Hicks, R. E., "Flow Patterns Upstream of Isokinetic Dust Sampling Probes," *Journal of the Air Pollution Control Association,* Vol. 28, June 1978, p. 601.

[4] Tine, G., *Gas Sampling and Chemical Analysis in Combustion Processes,* AGARDograph 47, Pergamon Press, Oxford, England, 1961.

[5] Clark, J. A., Moser, D. W., Best, W. D., and Thompson, N. A., "Temperature and Dilution Effects in Soot Mass Concentration Measurements," ASME Paper 83-GT-12, 1983.

[6] Dodds, W. J., Peters, J. E., Colket, M. B., and Mellor, A. M.,"Preliminary Study of Smoke Formed in the Combustion of Various Jet Fuels"*Journal of Energy,* Vol. I, March–April 1977, p. 119.

[7] Toone, B., "A Review of Aero Engine Smoke Emission," *Combustion in Advanced Gas Turbine Systems,* edited by I. E. Smith, Pergamon Press, Oxford, England, 1968, p. 276.

[8] Walker, C. L., "Particulate Measurements in Combustion," *Combustion Measurements,* edited by R. Goulard, Hemisphere, Washington, D. C., 1976.

[9] Kadota, T. and Henein, N. A., "Time-Resolved Soot Particulates in Diesel Spray Combustion," *Particulate Carbon: Formation During Combustion,* General Motors Research Labs., Warren, MI.

[10] Gillette, Allen, D., "Diesel Particulate Sampling and Analysis of Soot-Organic Interactions," M.S. Thesis, Purdue University, Mechanical Engineering, Dec. 1981.

[11] Dunleep, K. G., "Analysis of Automotive Particulate Sampling Technique," SAE Paper No. 800184, *Diesel Combustion and Emissions,* SAE 75th Congress and Exposition, Proceedings P-86, Detroit, MI, Feb. 25–29, 1980.

[12] Frisch, L. E., Johnson, J. H., and Leddy, D. G., "Effects of Fuel and Dilution Ratio on Diesel Particulate Emissions," SAE Paper 790417, 1979.

[13] MacDonald, J. S., Plee, S. L., D'Arey, J. B., and Schreck, R. M., "Experimental Measurements of the Independent Effects of Dilution Ratio and Filter Temperature on Diesel Exhaust Particulates Sampling," SAE Paper 800185, *Diesel Combustion and Emissions,* SAE 75th Congress and Exposition, Proceedings P-86, Detroit, MI, Feb. 25–29, 1980.

[14] Richards, G., Sojka, P. E., and Lefebvre, A. H., "Drop-Size Studies in a Radially-Uniform Fuel Spray," SAE Technical Paper Series 852083, 1985.

[15] Lefebvre, A. H., *Gas Turbine Combustion,* McGraw Hill, New York,1983.