The combustion of liquid fuel sprays is currently responsible for a large portion of the total energy consumption of the world. For example, most practical combustion systems, such as oil-fired base load power plants and home heating units, gas turbines, diesel engines, direct injected stratified charge engines, etc., operate using liquid fuel spray firing of the combustor. In these systems, a detailed understanding of the spray combustion phenomena is required in order to improve combustion efficiency and to predict and control pollutant formation and emissions. However, spray combustion depends upon and is governed by complex physical and chemical phenomena including simultaneous heat, mass, and momentum transfer, and the associated energy liberating chemical reactions. These factors are influenced by fuel type, droplet size distribution and number density, spray aerodynamics, ambient gas composition, and the local temperature and pressure. Because of these complexities and because of difficulties in direct measurement under actual operating conditions, the basic combustion processes, the rate of burning of fuel droplets, and the pollutant formation processes in spray combustion systems are not well understood at this time.

In view of its importance, the problem of spray burning has received considerable experimental and theoretical attention and has been the subject of several extensive reviews (e.g., Faeth, 1977, 1979; Chigier, 1975; Mizami, 1978; and others). Of particular interest and importance is the behavior of spray systems in the "transition region". In this region, which encompasses droplet sizes in the range of 25–80 \mu m diameter, the mixing and evaporation processes are both incomplete at the flame front and the burning occurs in a combined diffusive and premixed fashion. Under these conditions, the relative importance of heterogeneous and homogeneous effects in dominating the combustion process is switched and gives rise to a number of interesting and important effects. For example, maxima in burning velocities (Polymeropoulos and Das, 1975; Hayashi et al., 1976), extension of flammability limits (Burgoyne and Cohen, 1954), minima in ignition energies (Rao and Lefebvre, 1973; Ballal and Lefebvre, 1978; Cham and Polymeropoulos, 1981), and minima in pollutant emissions (Nizami and Cernansky, 1978, 1979; Nizami et al., 1982) have all been observed and reported. These transition region effects have been reviewed and discussed by Nizami et al. (1982). A brief overview of the behavior of oxides of nitrogen (NO\textsubscript{x}) emissions in the transition region is given below.

In their studies of NO\textsubscript{x} production in aerodynamically stabilized, 40–80 \mu m diameter, monodisperse isopropanol-air spray flames, Nizami and Cernansky (1978, 1979) observed significant droplet size effects in the transition region and an optimum droplet diameter with regard to lowest NO\textsubscript{x} levels. In general, as the droplet diameter in the spray decreases, NO\textsubscript{x} also decreases reaching a minimum value around 50 microns droplet diameter. Further reduction in droplet diameter increases the NO\textsubscript{x} ultimately reaching the constant premixed value. At an equivalence ratio of 1.39, a 61% reduction in NO\textsubscript{x} and a 87.5% reduction in NO were observed from the maximum values recorded in the two respective modes. These studies were extended later to include five additional hydrocarbon fuels with similar results (Nizami, 1978; Nizami et al., 1982). Typically, NO\textsubscript{x} reductions at the optimum droplet diameter were in the range of 40–70 percent and the optimum droplet diameter occurred from 48–55 microns. The occurrence of the minimum NO\textsubscript{x} point
at different droplet diameters for the different fuels appeared to be governed by the extent of prevaporation of the fuel in the spray, and was consistent with each fuel's physical properties. The minimum in NO\textsubscript{X} production as droplet size was varied was associated with droplet interaction effects and the transition from diffusive to premixed burning. Droplet interactions can result in a local temperature suppression with a subsequent reduction in NO\textsubscript{X} production; the transition point, and hence the significance of the droplet interaction effects, is governed by the extent of prevaporation.

While the experimental data and analysis were consistent for the above work, the general experimental configuration with an aerodynamically stabilized flame had undefined aerodynamic and mixing effects. Such effects could possibly be important and significant in biasing or affecting the data and conclusions. Thus, the basic experimental facility was modified to stabilize the flame on a screen flame holder and one dimensionalize the flow system, thereby eliminating these factors. This paper describes this modified experimental facility and reports new experimental data and results collected in the absence of mixing and aerodynamic effects on NO\textsubscript{X} formation in monodisperse spray combustion systems.

**EXPERIMENTAL APPARATUS AND PROCEDURE**

The basic experimental facility is shown schematically in Fig. 1. The facility included a Berglund-Liu Vibrating Orifice Monodisperse Aerosol Generator (Model 3050). This instrument, when operating in its monodisperse range, produces a uniform droplet stream with a standard deviation of less than 1% of the mean droplet size which, in turn, can be calculated to an accuracy of 2% or better from the generator operating conditions. To form a monodisperse aerosol, this stream of uniform droplets must be dispersed and diluted before significant coagulation occurs. This is accomplished by using dispersion and dilution air flows. The monodispersion ranges of the generator were established for a variety of orifices using the manufacturer's recommended "jet deflection" method and further confirmed by an impact slide and microphotograph techniques. The monodispersion ranges for the 20, 25, and 35 \textmu m orifice plates used in the present study are shown in Figure 2. Further details and characterization of the basic experimental facility are presented elsewhere (Nizami, 1978; Nizami and Germanusky, 1978).

After generation and dispersion, the spray of monodisperse droplets is supplied to the desired combustion system. In the original configuration used by Nizami and Germanusky (1979), the monodisperse fuel aerosols were supplied to and burned in an aerodynamically stabilized self-supporting flame inside a pyrex combustion tube. Primarily because of the uncontrolled mixing and aerodynamic effects in this configuration, as described earlier, the facility was modified to provide flame stabilization and one dimensional flow in the burner section. The combustion tubes were replaced with reducing and flow control sections directing the air/fuel aerosol to a 40 X 40 mesh stainless steel screen flame holder.
The new system includes: a water cooled flame holder to prevent uncontrolled fuel evaporation; a transition piece with geometrically streamlined curvatures; adjustable upper and lower housings to provide flexibility in burner height adjustment; and a baffle plate to improve and control the mixing of the dilution air with the dispersed aerosol. The current system configuration is shown in more detail in Figure 3.

The spray of monodisperse droplets was burned in a screen stabilized, one dimensional flame inside a pyrex chimney (ID = 32 mm, length = 267 mm). Fuel feed rate through the system was held constant at 0.382 cc/min. Measurements were made over the mono-dispersion operating range of the system encompassing droplet diameters from 36 to 70 um and equivalence ratios from 0.8 to 1.2. To change the equivalence ratio, the dilution air flowrate was varied keeping the dispersion air flowrate constant at 1300 cc/min. The ambient air temperature was 24°C for all the experiments. Flame holders were primarily selected to provide a stable flame and also to give approximately the same approach velocity at the screen flame holder. Thus, the combustor residence times were approximately the same at all conditions, and consequently, any residence time effects on NOx formation for the different operating conditions are expected to be insignificant.

Figurized and premixed conditions were examined as well to give the small droplet size limit. These prevaporized mixtures were made by mixing the fuel and dispersion air in a tee and then passing it through a vertical three-loop-helix wrapped with heating tape. The liquid fuel was vaporized and the mixture was heated to about 65°C before being introduced into the burner through the dispersion air path. This heating was sufficient to keep the mixture above the dew point of the fuel and to avoid condensation of fuel vapor on the combustor walls. Normal cooling of the mixture after leaving the vaporizer resulted in mixture temperatures near the flame holder of about 26°C.

Four liquid hydrocarbon fuels (isopropanol, methanol, n-octane, and n-heptane) were used for the experimental measurements. Post flame NO/NOx data were collected using an uncooled aerodynamically quenched stainless steel probe (0.30 mm orifice) at a fixed axial location of 5 cm above the screen flame holder.
Nitric oxide and total oxides of nitrogen were analyzed using a Thermoelectron chemiluminescent NO/NOx analyzer, modified for low pressure sampling. The accuracy of these measurements was \( \pm 0.5 \) ppm with \( \pm 1 \) ppm day to day reproducibility. Temperature measurements were made with a Pt/Pt-13% Rh fine wire thermocouple (0.003 inch/0.075 mm diameter) attached to the gas sampling probe. This combination temperature and gas sampling probe is shown schematically in Figure 4.

**EXPERIMENTAL RESULTS**

One dimensionality and symmetry of the flame for three different droplet sizes were examined by traversing the combination gas sampling/thermocouple probe across the combustion tube, 4 mm above the flame holder screen. The flame thickness for the 26.7 and 55.8 \( \mu \)m droplet sprays was about 3 mm and looked fairly flat. However, the flame shape for 69.1 \( \mu \)m droplets was extended to about 6 mm at the center. With the exception of 69.1 \( \mu \)m droplets, radial profiles of both NOx and temperature are indeed flat across the screen flame holder, as shown in Figures 5 and 6, thus confirming the one dimensionality of the burner system.

Axial traverses along the centerline of the burner were made as well. Representative temperature and NOx data for three droplet sizes and the associated premixed limit are shown in Figure 7. The data show the peaking of NOx and temperature in the flame zone with some reduction in the post flame region to nearly steady or slowly diminishing levels. The relative thickness of the flame zone is indicated in these types of plots. Based upon this information for the range of fuels being tested, a fixed sampling location on the burner centerline 5 cm above the screen flame holder was selected for fuel and droplet size comparison studies. This sampling location avoids regions of high temperature and concentration gradients.

Representative temperature and NOx results showing the effect of droplet diameter over a range of equivalence ratios are shown in Figures 8 through 11. Where available, premixed limit data are shown as data points at zero droplet diameter. For smaller droplet sizes, the flame was pale blue and nonluminous. Imbedded within the pale blue zone were smaller darker blue centers. The luminosity increased for heavier fuels. The thickness of the flame increased for larger droplet sizes, presumably because larger drops take longer to burn and thus, travel for a greater distance before burnout at the same flow velocity. A general trend, in these plots, is that NOx decreases as the droplet diameter is decreased reaching a minimum around 48-58 \( \mu \)m, and then increases to a fairly constant value with any further decrease in droplet diameter. Although the temperature and con-
concentration gradients are steep near the flame, this trend is also illustrated in the radial traverses, Fig. 6, where lower NOx levels are indicated with 55.8 μm than with 26.7 or 69.1 μm droplets. The small droplet size limit is generally close to and approaches the prevaporized premixed experiment results. The above mentioned general trend in NOx variation with droplet size was observed for all the fuels tested in rich, lean and stoichiometric burning situations. Also, it appears that, while the NOx minimum points are not sharply defined in the current data, the methanol minima are shifted to slightly larger droplet diameters and the n-heptane and n-octane data are shifted to slightly smaller droplet diameters relative to isopropanol. These general trends are in agreement with the theoretical calculations and the experimental data reported by Nizami et al. (1982) in their aerodynamically stabilized flame measurements. Thus, since aerodynamic and mixing effects have been minimized in the current system, these results and behavior indicate a true droplet size effect on NOx formation.

DISCUSSION

The results just presented indicate that relative to larger droplet diameters (>60 microns), where diffusive type of burning dominates, NO and NOx decrease with decreasing droplet diameter. These results are in agreement with the theoretical and experimental studies of Bracco (1973), Kesten (1972), Labowski (1980), and others. For individual droplet burning, decreasing the droplet size increases the flame to droplet diameter ratio, reduces the flame temperature, and therefore, results in reduced NOx production. In contrast to individual droplet combustion, spray burning results in droplet interactions which decrease the local oxygen concentration around burning droplets, resulting in a subsequent reduction of flame temperature and NOx.

At smaller droplet diameters (<30 microns), where premixed type of burning dominates, NO and NOx remain relatively constant with decreasing droplet diameter. In this regime, sufficient fuel is prevaporized and premixed prior to the flame front so that the situation approaches the premixed case.

In the transition region (30-70 microns), where the NOx minimum is observed, the combustion undergoes a transition from diffusive to premixed burning domination of the system. As the droplet size is reduced, additional fuel is prevaporized until the situation resembles vaporized fuel seeded with small liquid droplets, which eventually approaches the premixed case. As this transition takes place, relatively less oxygen depletion occurs, and therefore, the temperature and NOx go through minimum and gradually increase to their prevaporized and premixed levels.

The apparent shifts in the droplet diameters associated with the NOx minimum points for the various fuels were found to be consistent with the thermophysical properties of the fuels; some of these thermophysical properties and evaporation parameters, as reported by Nizami et al. (1982), are given in Table 1. The evaporation constants and extent of prevaporization for the different fuels were calculated and compared following the approach used by Nizami et al. (1982). The procedure yields reasonable relative results for extent of prevaporization prior to the fuel spray reaching the flame zone. Using isopropanol as the base case, the mass ratios of fuel evaporated relative to isopropanol for fixed initial droplet diameters were calculated. The mass ratio for methanol was larger than one and the mass ratios for n-heptane and n-octane were less than one. Thus, methanol with its higher evaporation ratio, requires larger droplets to produce the same levels of prevaporization as the isopropanol.
FIGURE 8. DROPLET SIZE EFFECT ON POST FLAME TEMPERATURE, NO, AND NOx FORMATION IN MONODISPERSE AEROSOL COMBUSTION FOR VARIOUS EQUIVALENCE RATIOS AT A FUEL FEED RATE OF 0.382 CC/MIN USING ISOPROPANOL AS FUEL. O - NO; • - NOx; △ - TEMPERATURE.

FIGURE 9. DROPLET SIZE EFFECT ON POST FLAME TEMPERATURE, NO, AND NOx FORMATION IN MONODISPERSE AEROSOL COMBUSTION FOR VARIOUS EQUIVALENCE RATIOS AT A FUEL FEED RATE OF 0.382 CC/MIN USING N-HETANE AS FUEL. O - NO; • - NOx; △ - TEMPERATURE.
FIGURE 10. DROPLET SIZE EFFECT ON POST FLAME TEMPERATURE, NO, AND NOX FORMATION IN MONODISPERSE AEROSOL COMBUSTION FOR VARIOUS EQUIVALENCE RATIOS AT A FUEL FEED RATE OF 0.382 CC/MIN USING METHANOL AS FUEL. ○ - NO; ● - NOX; Δ - TEMPERATURE.

FIGURE 11. DROPLET SIZE EFFECT ON POST FLAME TEMPERATURE, NO, AND NOX FORMATION IN MONODISPERSE AEROSOL COMBUSTION FOR VARIOUS EQUIVALENCE RATIOS AT A FUEL FEED RATE OF 0.382 CC/MIN USING N-OCTANE AS FUEL. ○ - NO; ● - NOX; Δ - TEMPERATURE.
base case. N-octane and n-heptane, on the other hand, require smaller droplets to achieve the same levels of prevaporization. Thus, these calculations are consistent with the experimental data and indicate the importance of the extent of prevaporization in determining the NO$_x$ minimum point.

Nizami et al. (1982) have also suggested that single component heavy fuels are expected to shift the minimum NO$_x$ point to lower droplet diameters than 50 microns. Further, in the case of multicomponent heavy fuels, the behavior is expected to be governed by the lighter components because they will determine the prevaporization characteristics of the fuel. In a polydisperse spray of a heavy multicomponent fuel, similar trends are expected. However, additional experimental work will be required in order to elucidate the NO$_x$ behavior in practical combustion systems, which have different pressure and temperature environments, and widely varying droplet sizes.

CONCLUSIONS

A one dimensional aerosol spray combustion facility has been developed and characterized. Experimental results of post flame NO/NO$_x$ emissions utilizing liquid hydrocarbon fuels in this combustion system have been presented. A significant droplet size effect on NO/NO$_x$ emissions was observed for the four fuels tested. Both NO and NO$_x$ decrease with decreasing droplet diameter, reach minima (around 48-58 μm droplet diameter), and then increase again with further decreases in droplet size, eventually approaching the premixed combustion limit. Further, the minimum NO/NO$_x$ point shifts towards a smaller droplet size for lower vapor pressure single component fuels.

Overall, the observed effects are attributed to droplet interactions and the transition from diffusive to prevaporized and premixed burning. Droplet interactions apparently result in local temperature suppression with a subsequent reduction in NO$_x$ production. The observed occurrence of the minimum NO$_x$ point at different droplet diameters for individual fuels is explained through theoretical calculations of the extent of prevaporization of the fuel in the spray. The theoretical analysis and experimental evidence indicate that the extent of prevaporization is an important factor in determining the minimum NO$_x$ point.

This study confirms the existence of a droplet size dependence on NO$_x$ emissions in the transition region in the absence of mixing and aerodynamic effects. Consequently, reduced NO$_x$ production should be obtainable through appropriate droplet size control. This fact (and the associated experimental data) may be useful in designing and optimizing hydrocarbon fuel spray combustion systems.

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REFERENCES


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<td>Isopropanol</td>
<td>C$_3$H$_8$O</td>
<td>60.10</td>
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<td>65.95</td>
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<td>87.1</td>
<td>19.26</td>
<td>0.089</td>
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<tr>
<td>Methanol</td>
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<td>263.0</td>
<td>231.04</td>
<td>0.482</td>
<td>0.963</td>
<td>3.608</td>
</tr>
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*Nizami et al. (1982)


