# Effect of Turbulence on Pollutant Formation in a Tubular Burner

G. F. Robinson*
*Clemson University, Clemson, S. C.*
and
A. A. Kovitz†
*Northwestern University, Evanston, Ill.*

**The role of turbulence in pollutant formation was investigated both experimentally and analytically with the objective of understanding the important mechanisms on a fairly basic level. Preliminary measurements of the cold-flow turbulence characteristics downstream of three perforated plate flame holders, of varying solidity ratio, were made. In the combustion experiment, measurements of the concentration distributions of CO, $NO_x$, HC, and $CO_2$ downstream of the same flame holders were made. The combustor configuration was tubular and of constant area, and the fuel was premixed, prevaporized propane. The experimental results were modeled by a one-dimensional analysis in which transport phenomena were neglected and, more successfully, by a quasi-one-dimensional analysis containing recirculation zones and turbulent transport effects. It was concluded from the analysis that the CO oxidation rate was mixing limited, but that this rate could be increased by increasing flame holder solidity and hence turbulent mixing rate. The $NO_x$ production was governed by its formation in the recirculation zones and turbulent transport into the mainstream flow. A method for reduction of combustor emissions utilizing these effects is suggested.**

## Introduction

FORECASTS in recent years indicate that the gas turbine engine, as a result of its development for aircraft use, can be expected to play an increasing role in the utilization of fossil fuels over the next several decades.[1] It is therefore a worthy object of research attention. In the present study we have focused our attention on the air-polluting aspects of these engines, and attempted to elucidate these aspects by examining the basic pollutant formation mechanism in a burner of fairly simple configuration. Furthermore, since the interaction between fluid mechanics and combustion is an area in which there is ample room for an increase of understanding,[2] it was felt that the present effort should be made in that direction. In this study, one aspect of fluid mechanics, turbulence, was chosen for further scrutiny as to its role in pollutant formation.

There has been a good deal of previous work in the related areas of turbulent flames, chemical kinetics, and pollutant formation by combustion. We have attempted to take this work into account in so far as possible. Turbulent flame research was characterized in the early years by measurements of the flame speed and analyses of the geometrical configuration of the flame.[3-7] This work has continued over the years and has been marked by controversy and lack of agreement. Thus, the understanding of these aspects of turbulent flames has not progressed sufficiently to be of much use in analyzing real combustor systems. Turbulent flame research has also investigated the question of flame-generated turbulence, but it is now felt, as a result of experimental and analytical investigations, that this quantity is very small, if present at all.[8-10]

In recent years turbulent flame research has also focused on the flames occurring in real combustion systems. In this vein, various aspects of recirculating flow have been considered in their relation to combustion. Highlighting this work has been the development of numerical methods whereby the partial differential equations describing such flows may be solved directly.[11] Turbulent flames in real combustion systems have also been described by the perfectly stirred reactor concept and, in this regard, analyses and experiments have been performed to describe the effect of deviations from the ideal of perfect stirring.[12-14] In the most recent developments, the statistical theory of turbulent combustion has been considered in an analogous way to the theory of turbulence in non-reacting flows.[15-18]

Much work has also been done in studying the chemical kinetics of combustion. Through this basic research, the mechanism and rate constants of hydrogen-oxygen combustion are now fairly well determined. Unfortunately, the same cannot yet be said about even the simplest hydrocarbon flames. Although the mechanism of methane flames is understood to some limited extent, those of propane and the higher hydrocarbon flames are understood much less. Even in cases where the individual reaction steps have been identified with some certainty the rate constants are still very uncertain.[19] For this reason measured overall combustion rates are still used to a great extent in design and analysis.[20] One important conclusion has been reached, however, in regard to the kinetics of hydrocarbon combustion, and this is that the oxidation of CO is the rate-limiting step in the sequence of elementary reactions. Consequently, this step has received considerable research attention to obtain better data on its rate.[21,22]

The chemical kinetics of nitric oxide formation in flames has also received much attention of late. There is general agreement that the reaction of $N_2$ with monatomic oxygen is the principal formation step. Much less certain is a possible mechanism for the formation of "prompt NO" in fuel-rich flames.[23-25]

Research on the formation of pollutants in gas turbine engines has had a shorter history than that of basic research on turbulent flames and the chemical kinetics of combustion. This work has been characterized by the measurement of pollutants in the exhaust of actual aircraft engines,[26-28] experiments in model combustors in an attempt to determine the controlling factors,[29,30] analytical studies of pollutant for-

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*Assistant Professor of Mechanical Engineering. Member AIAA.

†Professor of Mechanical Engineering and Astronautical Sciences. Member AIAA.

mation with particular attention to the role of mixing,[31,32] analytical modeling of real gas turbine combustors,[33-35] and experimentation with proposed methods of reducing pollutants.[36]

## Present Study

The present study consisted of an experimental investigation conducted at the NASA Lewis Research Center, followed by an analytical modeling of the experimental results. A detailed report of this work was made previously.[37] In the experimental phase, preliminary measurements of the cold-flow turbulence characteristics downstream of three perforated-plate flame holders, of varying solidity ratio, were made. This was followed by the measurement of the concentration distributions of CO, $NO_x$, HC, and $CO_2$ behind the same flame holders with combustion at three different equivalence ratios. The preliminary cold-flow measurements were made to obtain quantitative information on the turbulence characteristics, which was then used to infer the turbulence properties in the combusting flow.

The configuration of the combustor used for these measurements is illustrated in Fig. 1. The combustor was tubular, of constant area, and constructed from pipe of 10.2-cm diam. Evaporated propane was premixed with the airstream at a location of 1.95 m upstream of the flame holder. Gas samples were drawn continuously from the combustor by a traversing probe located on the axial centerline. To ensure structural integrity of the combustor it was provided with a cooling water jacket, and a water spray was used to quench the exhaust gases and thus protect the downstream piping and back-pressure valve (not shown). There was no addition of dilution air in the combustor.

## Turbulence Measurements

The cold-flow turbulence measurements were made in a test section identical geometrically with the combustor. Upstream of the test section the airflow was smoothed, in the conventional way, by using a "honeycomb" flow straightener, a section packed with steel wool, quieting screens, and a diameter contraction. The perforated plates used to generate turbulence were identical with the flame-holder plates used in the combustion experiment. They were 0.635 cm thick, with 0.635-cm diam holes arranged axisymmetrically. Solidity ratios were 0.742, 0.815, and 0.892; nominal mesh lengths were 1.12 cm, 1.32 cm, and 1.70 cm. Experimental conditions for the turbulence measurements consisted of a constant airflow rate of 200 g/sec, a velocity of 21.3 m/sec, and a duct Reynolds number of 130,000. The air temperature was ambient and the test section was at atmospheric pressure.

The experimental measurements consisted chiefly of the airflow rate, the properties upstream of the perforated plate, the plate differential pressure, and the time-mean and fluctuating velocity at a large number of axial and radial positions within the test section. The latter measurements were made using hot-wire anemometry. Measurements of secondary interest were the autocorrelation coefficient, from which the scale of turbulence was computed, the turbulence energy spectrum, and the plate pressure-coefficient.

The experimental observations of the time-mean velocity distribution indicated the following points:

1) The velocity decreased with increasing distance from the plate, due to the reverse pressure gradient.

2) The radial distribution of the axial velocity component, at axial distances near the plate, was strongly dependent on the hole pattern, i.e., the distribution of local solidity ratio. For two of the plates used, this resulted in an off-centerline velocity peak. There were indications, however, that axial symmetry of the velocity profile was maintained.

3) The radial distribution of the axial velocity component became flatter with increasing distance from the plate.

4) A relatively large recirculation zone was located in an outer annular ring immediately behind the plate, due to the 100% solidity of the plate in this area.

The experimental observations of the turbulence intensity distribution indicated the following points:

1) Within the first few centimeters downstream of the plate, relatively low turbulence intensities were observed in the center of the duct, but the turbulence intensity rose sharply near the wall.

2) Farther downstream, the centerline turbulence intensity rose to a peak, the radial distribution flattened, and then a general decay of the turbulence intensity was observed.

3) The axial variation of turbulence intensity had a peak value about 15 cm from the plate, when considering a radially averaged turbulence intensity to be representative of each cross-section of the flow. These peak values were 36%, 46%, and 51% for the three plates used. The data are shown in Fig. 2, where the abscissa is expressed as the number of hole diameters from the plate.

4) The decay of turbulence followed a linear law for $(U/u')^2$ vs $(x/M)$, as has been observed by others for the initial period of decay.

5) The decay of turbulence was also correlated well by the Swithenbank turbulence model[38] in which a turbulence energy balance concept is used. In this correlation, the eddy-size parameter was 1.4 (multiples of mesh length) and the mixing length parameter was 0.02 (multiples of mesh length).

The experimental observations of turbulence scale indicated that the data on all three plates were well correlated with each other when nondimensionalized against the mesh length. Values of the scale ranged from 1.3 to 1.5 times the mesh length, except immediately behind the plate.

## Combustion Measurements

The principal measurements in the combustion experiment were the measurements of the CO, $NO_x$, HC, and $CO_2$ concentrations along the axial centerline of the combustor. These were made by continuous sampling of the combustion gases through a traversing probe fabricated with concentric, stainless-steel, tube passages. Cooling water, for quenching the sample reactions, flowed through two concentric annular passages; the sampled gas flowed through a central passage. Outside diameter of the sampling probe was 1.27 cm. The gas

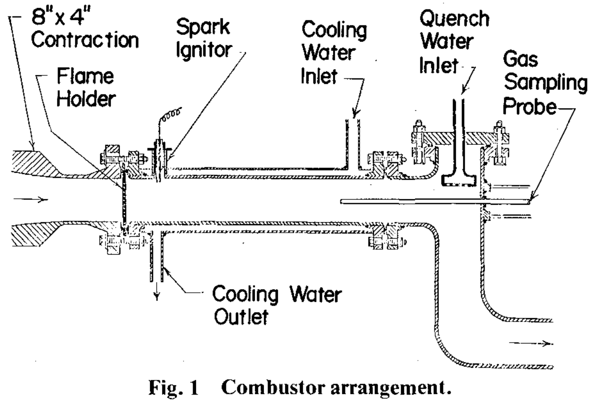

Fig. 1 Combustor arrangement.

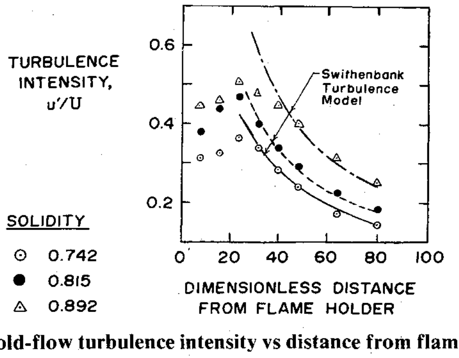

Fig. 2 Cold-flow turbulence intensity vs distance from flame-holder.

analysis instrumentation consisted of nondispersive infrared analyzers for CO and $CO_2$, a flame ionization detector for HC, and a chemilluminescent analyzer for $NO_x$. The oxides of nitrogen were measured on a total $NO_x$ basis, although periodic checks indicated that oxides other than NO were present in only negligible amounts. No attempt at direct measurement of combustor temperatures was made, but inferred temperatures were computed from the species concentration measurements.

*Variable* experimental conditions consisted of the three flame-holder solidity ratios and the three equivalence ratios (0.635, 0.730, 0.845). *Fixed* experimental conditions included mass-flow rate (640 g/sec), inlet temperature (590 K), combustor pressure (5 atm), and exhaust-gas Mach number (0.1).

The results of the species concentration measurements for CO, $CO_2$, and $NO_x$, at an equivalence ratio of 0.635, are shown in Figs. 3-5. On each of these plots the data for the different flame holders are compared. The dimensionless distance variable, on each of these plots, is expressed in terms of the number of flame-holder hole diameters.

The experimental observations of the CO concentration distribution indicate the following points: 1) The CO concentration decreased with increasing distance from the flame holder, due to the oxidation of CO to $CO_2$. Equilibrium of the CO level was reached before the combustor outlet in most cases. 2) The peak concentration of CO in the combustor occurred within the first 4 cm downstream of the flame holder, a region in which measurements were not made. 3) There was a trend toward more rapid oxidation of CO as the flame-holder solidity ratio was increased. This was most pronounced at the lowest equivalence ratio, and gradually faded out as the equivalence ratio was increased. 4) A possibility of imperfect quenching of the gas sample was indicated at the highest equivalence ratio.

The experimental observations of the $NO_x$ concentration distribution indicated the following points: 1) A steady rise in $NO_x$ concentration with increasing distance from the flame holder was observed. 2) Within the limits of the experimental data scatter, no variation in $NO_x$ formation rate with flame-holder solidity ratio could be observed. 3) The $NO_x$ formation rate was much slower than that required to produce equilibrium concentrations at the combustor outlet. 4) A strong effect of temperature on the $NO_x$ formation rate was observed in comparing the data at different equivalence ratios.

The experimental observations of the HC concentration distribution indicated the following points: 1) The HC concentration signal from the flame ionization detector showed a high degree of intermittency, and this resulted in a good deal of data scatter. 2) The trends in the HC concentration curves, with increasing solidity ratio and equivalence ratio, paralleled those of the CO concentration curves. 3) The presence of relatively large amounts of HC during certain experimental conditions was taken to be an indication of mixing-limited combustion.

The plots of a reaction-completeness variable and the inferred combustor temperature were also useful, since they indicated, in a composite way, the effect of solidity ratio and equivalence ratio on the combustion rate. These plots also emphasized that the gas sampling was being done only at the tail end of the combustion process.

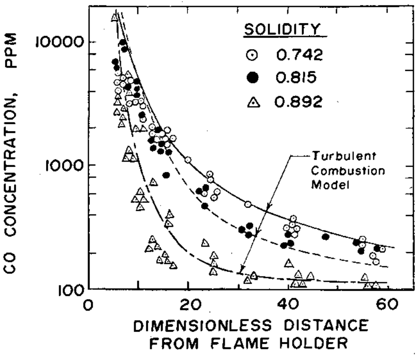

Fig. 3 CO concentration vs distance from flame holder.

## Analytical Modeling

Following the experimental work just described an analytical study was undertaken to assist in the interpretation of the data. As an initial analytical approach, the computations of a Rankine-Hugoniot analysis were made and compared to the data. This type of an analysis does not describe the detailed property variations within a flame, and so is of very limited usefulness in pollutant formation studies. Nevertheless, its use was beneficial in developing computational tools, particularly those for computing chemical equilibrium compositions, which were used in the later analyses. In comparing the results of the Rankine-Hugoniot analysis to the experimental data, we were able to evaluate the approach of the species concentrations to the equilibrium values. It was thus seen that the CO attained equilibrium at the combustor outlet, but that the $NO_x$ was still far below the equilibrium level of nitric oxide. Increased equivalence ratio, however, enabled the $NO_x$ to approach more closely to the equilibrium level.

Following the Rankine-Hugoniot analysis, more sophisticated analytical approaches were considered. One of these approaches is that in which the assumptions of a one-step chemical reaction and a Lewis number of unity are made.[39] This method was not applied in the present study, however, because the many simplifying assumptions make it ineffective in pollution research.

The various methods of analysis which include chain-reaction chemistry and radical distributions were also considered.[39] As an application of these methods, computations were made with an analysis in which the transport phenomena were neglected (plug flow).

Formulation of this analysis followed from the general one-dimensional conservation laws for gas mixtures flowing in a duct. With the neglect of transport phenomena, the species continuity equations reduced to first-order ordinary differential equations relating the time rate of change of species concentration to the chemical reaction rates. Additional relations consisted of the energy equation, equation of state, and the phenomenological chemical kinetic rate expression, all of which were algebraic. The assumed chemical mechanism

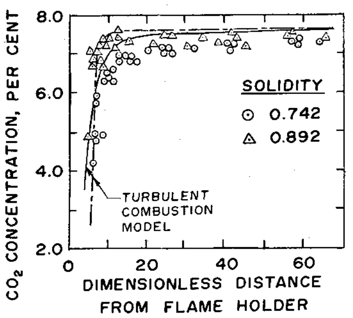

Fig. 4 $CO_2$ concentration vs distance from flame holder.

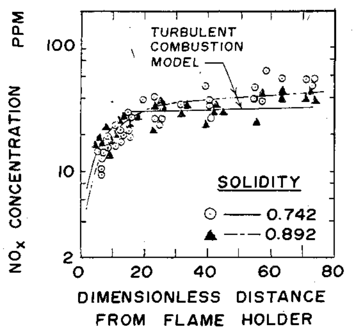

Fig. 5 $NO_x$ concentration vs distance from flame holder.

consisted of 25 elementary reaction steps involving 12 chemical species. Oxides of nitrogen were modeled entirely as nitric oxide. The solution of the governing differential equations was facilitated by use of a recently developed numerical integration scheme for "stiff" differential equations.[40]

The result of the plug-flow analysis overpredicted the CO oxidation rate in those cases for which mixing limitations were believed to be controlling. Furthermore, the $NO_x$ formation rate was underpredicted by the analysis, seemingly because the formation of $NO_x$ in the recirculation zones was not taken into account. It was, therefore, concluded that a one-dimensional analysis was inadequate because it did not admit the possibility of nonhomogeneities in composition or the presence of recirculation zones.

As a result of these conclusions, a quasi-one-dimensional turbulent combustion model was formulated to account for both the nonhomogeneities and the recirculation zones, as well as turbulent mass transport. In previous studies, recirculation zones were considered when modeling combustion,[41] and this work has been taken into account in the present case. The general characteristics of the present model were derived from a consideration of the actual flow pattern in the combustor, as depicted schematically in Fig. 6.

Since the experimental flames were stabilized on perforated-plate flame holders, the flow pattern was characterized by 1) high-velocity jets issuing from each hole, and 2) recirculation zones behind the solid portions of the plate. The individual jets expanded in size until coalescence of the several jets occurred at some point downstream of the plate. The size of the recirculation zones gradually decreased until they disappeared completely at the same point. Thus, in the model, the flowfield was divided into jet-flow zones and recirculation zones, and certain characteristics were assumed for each. The boundary surfaces separating the two types of zones were chosen to coincide with the limiting streamlines of the flow passing through the plate holes. Therefore, there was no net mass flow across the dividing surfaces, although there was turbulent transport of individual species in one direction or the other.

The jet flow zones were assumed to contain a one-dimensional, variable-area flow of a 12-component mixture undergoing chemical reaction. A multistep reaction mechanism, identical with that used with the plug-flow analysis, was initiated by a step of infinite speed which described the actual initial reaction steps in a global way. The physical location of this initiation, that is, its distance from the flame-holder plate, was a parameter of the model ($K_0$). Within any given cross-section of a jet-flow zone it was assumed that nonhomogeneities in composition could exist due to the finite rate of mixing between the jet-flow zone and the surrounding recirculation zone. As a result of this, the overall combustion rate was sometimes limited by the mixing rate rather than by the chemical reaction rates.

To account for this effect in the model, the chemical reaction rate of each elementary step was multiplied by a mixedness parameter ($K_1$) of value less than or equal to unity. That is, if the mixing rate was very fast, the mixedness parameter was set equal to unity, and the combustion rate was governed only by the chemical reaction rates. If the mixing rate was slow, the mixedness parameter was set equal to a value less than unity, and the chemical rates were restrained by the mixing limitations.

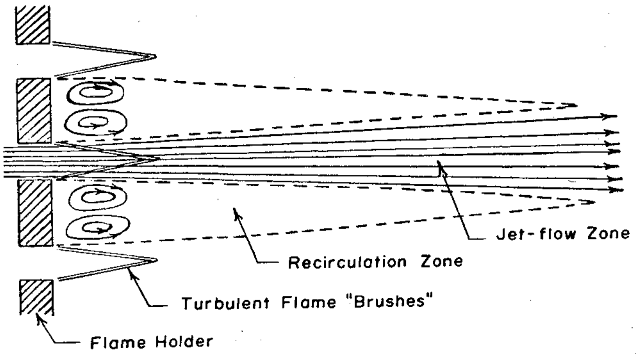

Fig. 6 Schematic representation of combustor flow.

In the recirculation zones, it was assumed that the residence time was long compared to the characteristic chemical reaction time for combustion. Therefore, it was assumed that each recirculation zone was a homogeneous zone containing equilibrium concentrations of all species (except NO) at the adiabatic flame temperature. Because of the slow formation kinetics of NO, the time necessary for its equilibration was, however, much longer than the assumed residence time in the recirculation zones. Therefore, the ratio of the recirculation-zone NO concentration to the equilibrium NO concentration was another parameter ($K_2$) of the model.

The recirculation zones were assumed to be in steady state. With regard to mass flow, a steady-state condition was ensured by judicious choice of the zone boundaries, as mentioned earlier. To ensure steady-state energy flow, the various energy transfers were set in balance, locally, across each element of the zone boundary surface. In essence, this meant that the net flux of energy from the recirculation zones to the jet-flow zones by thermal conduction and turbulent mass transport was in balance with a net flux of ehemical energy in the opposite direction.

This model was formulated in terms of the conservation laws for mass, species, and energy, as well as necessary subsidiary relations, such as the phenomenological chemical kinetic rate expression, ideal gas equation of state, and caloric equation of state. In addition to these equations, relations were introduced to express the turbulent mass transport rates in terms of the pertinent variables. In brief, the principal equations of the analysis were the 12 species continuity equations

$$dY_i/d\zeta = (3/2)\Omega_i (r/r_0)^2 + 0.112(\rho/\rho^*)^{-1}(r/r_0)^{-1}(Y_i^* - Y_i)$$

This equation expresses the rate of change of species mass fraction $Y_i$ with dimensionless distance from flame holder $\zeta$ as a function of dimensionless species production rate $\Omega_i$, dimensionless jet-zone radius $r/r_0$, dimensionless jet-zone density $\rho/\rho^*$, and the recirculation zone mass fraction $Y_i^*$. The first term on the right-hand side of the equation contains the effect of chemical reaction, while the second term contains that of turbulent mass transport between the jet-flow zones and the recirculation zones. Complete details of the analytical treatment are given in the report cited.[37]

The effect of the parameter values on the model predictions of polluting species concentrations was of special interest. In this regard, it was found that the flame location parameter $K_0$

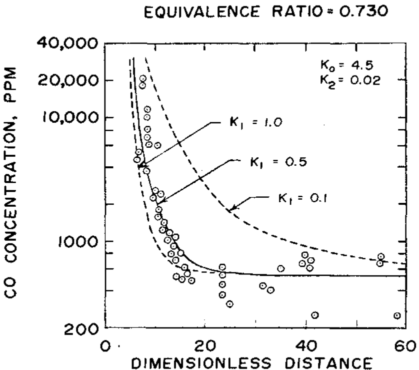

Fig. 7 Effect of mixedness parameter.

did not have a significant effect on the predicted distributions of either the CO or NO. The mixedness parameter $K_1$ had a strong effect on the CO predictions, as shown in Fig. 7, but a negligible effect on the NO predictions. The recirculation-zone NO concentration parameter $K_2$ had a strong effect on the NO predictions, as shown in Fig. 8, but a negligible effect on the CO predictions. As a result of this, the CO and $NO_x$ data were used to evaluate the parameters by determining the values which provided the best fit between the model predictions and the experimental data. These parameter values were found to be dependent on both the flame-holder solidity ratio and the equivalence ratio, but in a manner that could be readily interpreted on physical grounds. This interpretation, in turn, provided additional insight into the physical phenomena which had been observed, and is presented in the conclusions given later.

Since the parameters had been evaluated by consideration of the CO and $NO_x$ data, an independent check of the model was provided by comparing the model predictions of both the $CO_2$ concentration and the temperature to the experimental data on these variables. The results of this check were relatively good.

The overall output of the model, in regard to predictions of species concentrations, is shown in Fig. 9. For the computation shown, the chemical reactions were initiated at a distance $\zeta$ of 4.5 from the flame holder. At this point, there is a discontinuity in the concentration curves as a result of the infinitely fast initiation step of the chemical reaction mechanism. In accordance with this, the $C_3H_8$ is entirely consumed, the $O_2$ is partially consumed, and a considerable amount of CO and $H_2O$ is produced. For $\zeta<4.5$, this plot shows the effect of turbulent mass exchange between the two zones. For $\zeta>4.5$, both the effects of turbulent mass exchange and chemical reaction are present. The relative sizes of the chemical reaction and turbulent mass-exchange terms are shown in Figs. 10 and 11. In these figures, it is seen that chemical reaction dominates in the production of $CO_2$ (and destruction of CO), but that turbulent mass-exchange dominates in the production of NO.

## Conclusions

The turbulent combustion model was able to correlate the experimental data quite well and, together with the plug-flow analysis, enabled us to draw several conclusions on the pollutant-forming role of turbulent transport and recirculation zones in the experimental burner. These conclusions are summarized as follows:

1) Local nonhomogeneities in the mainstream, or jet-flow zones (as characterized by the $K_1$ parameter), are created by the presence of recirculation zones behind a bluff-body flame stabilizer, even though the fuel and air may be thoroughly premixed upstream.

2) These nonhomogeneities in the jet-flow zones are reduced as the flame-holder solidity ratio, and hence the turbulence level and mixing rates, are increased. This effect is, therefore, a result of increased turbulent transport *within* the jet-flow zones.

3) These nonhomogeneities in composition are rate-limiting in the oxidation of CO to $CO_2$. Thus, the oxidation rate is increased by increasing the flame-holder solidity ratio.

4) This rate-limiting effect of the nonhomogeneities on CO oxidation rate becomes less pronounced as the equivalence ratio is increased away from the lean blowout limit.

5) Overall $NO_x$ production is dominated by a) the formation of $NO_x$ in the recirculation zones behind bluff-body flame stabilizers, and b) the rate at which this $NO_x$ is transported, via the turbulent transport mechanism, *across the boundaries* of the recirculation zones into the jet-flow zones. In the analytical model, this transport mechanism is considered separately from that mentioned above in item 2.

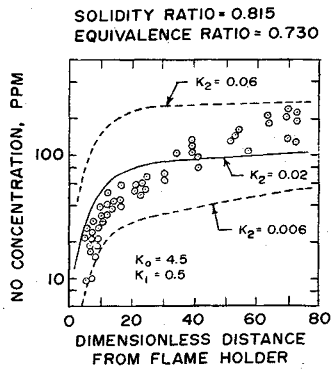

**Fig. 8 Effect of recirculation-zone NO concentration parameter.**

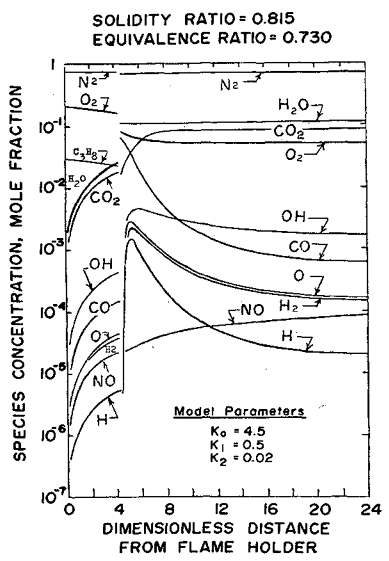

**Fig. 9 Species concentrations in turbulent combustion model.**

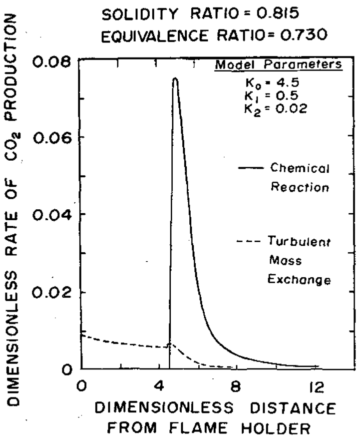

**Fig. 10 Rate of $CO_2$ production in jet-zones of turbulent combustion model.**

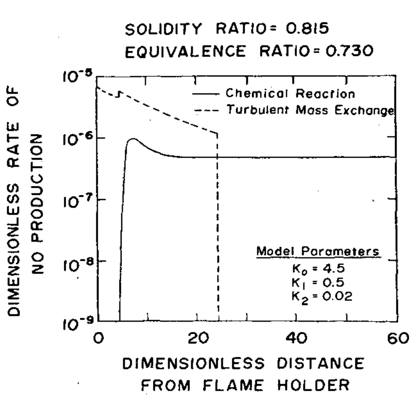

**Fig. 11 Rate of NO production in jet-zones of turbulent combustion model.**

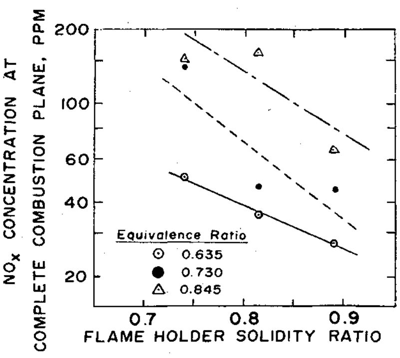

**Fig. 12 $NO_x$ concentration at "complete combustion" plane.**

6) Increasing the flame-holder solidity ratio shortens the combustor length required for complete CO oxidation (via items 2 and 3), and therefore may be used to reduce $NO_x$ emissions from a real gas turbine combustor by permitting earlier introduction of dilution air, and hence earlier quenching of the $NO_x$ formation reactions.

The last conclusion is illustrated by Fig. 12 in which we have plotted the measured $NO_x$ concentration at the "complete combustion" plane (i.e., the combustor plane at which CO oxidation is essentially complete) vs the flame-holder solidity.

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