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AN EXPERIMENTAL STUDY OF
REACTIVE TURBULENT MIXING

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ABSTRACT

An experimental study of the turbulent mixing of two coaxial gas streams, which react very rapidly (pseudo-hypergolic chemical kinetics), was performed to investigate the mixing characteristics of turbulent flow fields. The center stream consisted of a CO-N₂ mixture and the outer annular stream consisted of air vitiated by H₂ combustion. The streams were at equal velocity (50 m/sec) and temperature (1280 K). Turbulence measurements were obtained using hot film anemometry. A sampling probe was used to obtain time averaged gas compositions. Six different turbulence generators were placed in the annular passage to alter the flow field mixing characteristics. The turbulence generators affected the bulk mixing of the streams and the extent of CO conversion to different degrees. The effects can be related to the average eddy size (integral scale) and the bulk mixing. Higher extents of conversion of CO to CO₂ were found by increasing the bulk mixing and decreasing the average eddy size.

INTRODUCTION

An experimental study of the turbulent mixing of two coaxial streams which react very rapidly (pseudo-hypergolic chemical kinetics) was performed to investigate the mixing characteristics of turbulent flow fields. The effects of varying the flow field mixing characteristics upon the bulk mixing and the extent of conversion of a chemical tracer were studied.

Reaction times in high intensity combustors are mixing dependent. The rate of formation of oxides of nitrogen is reduced by rapid quenching of the gases leaving the primary zone. Carbon monoxide burn up is very dependent upon the mixing of combustion gases and air in the secondary zone. A knowledge of the turbulent mixing processes is very important to the prediction of pollutant formation and combustion efficiency (refs. 1-4). However, little data are available relating turbulent intensity and scale to the degree of molecular mixedness or chemical conversion. Keeler, et al (ref. 5), and Vassilatos and Toor (ref. 6) have utilized very fast, acid-base neutralization reactions for the study of turbulent-chemical interactions in one dimensional flow experiments. Singh (ref. 7) has studied the coaxial mixing problem also using acid base reactions. Batt, et al (ref. 8) have investigated gaseous pseudo-hypergolic turbulent mixing in a two-dimensional shear layer. A nitrogen dioxide reaction was used to follow the mixing processes.

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The research described in this report was performed to provide detailed information on turbulence characteristics and chemical conversion for gaseous coaxial mixing. This type of flow resembles that of gas turbine combustors more closely than previous experiments.

A pseudo-hypergolic reaction scheme was selected for this research because the extent of conversion is mixing limited and independent of the chemical kinetics. The turbulent mixing field can be studied directly by observing the degree of reaction completeness. This type of experiment is superior to a diffusion flame since the species react on contact at the molecular level and no piloting is required to sustain the reaction. The measured properties thus reflect the turbulent chemical interaction of the entire flow field rather than the behavior of a pilot zone.

The experimental study was carried out in a confined coaxial flow configuration consisting of a center jet of carbon monoxide and nitrogen mixing with an outer annular flow of air which had been vitiated by hydrogen combustion. The reactant streams were both preheated to a temperature of 1280 K to achieve pseudo-hypergolic reaction rates. Mixing characteristics were altered by placing six different turbulence generators in the outer annular stream.

Measurements of the time averaged species mole fractions, temperatures and velocity were obtained. The extent of reaction completion was obtained using a specially designed, water cooled, gas sample probe. This probe incorporated a supersonic convergent-divergent nozzle to quench the chemistry rapidly.

The turbulent flow field was characterized by measuring mean velocity, axial turbulent intensity, dissipation and integral scales at ambient pressure and temperature. The flow rates used in the cold flow experiments were selected to achieve Reynolds number similarity with the high temperature experiments.

**SYMBOLS**

- $D_0$: diameter of center stream pipe, 2.66 cm
- $M$: mesh length, cm
- $r$: radial distance from center line, cm
- $R_o$: radius of center stream pipe, 1.33 cm
- $U'_\text{LOCAL}$: point rms value of the fluctuating velocity, m/sec
- $U_{\text{LOCAL}}$: time averaged point velocity, m/sec
- $U_{\text{AVG}}$: time and spacial averaged bulk velocity, m/sec
- $X$: axial distance from centerstream exit plane, cm
- $A$: integral scale of turbulence, cm
APPARATUS AND PROCEDURE

Chemical System

The primary requirement for this study was a chemical system which was mixing limited and independent of the chemical kinetics. A carbon monoxide-oxygen-water vapor chemical system was selected on the basis of facilities, safety, and compatibility with on-line gas sampling procedures. In order to approach infinitely fast kinetics with this chemical system, preheating of the reactants was required. Preheating was accomplished by burning hydrogen in the air stream. This also introduced into the chemical system, water vapor which catalyzes the carbon monoxide oxidation.

The overall temperature rise due to the oxidation was limited by greatly diluting the carbon monoxide stream with N₂. The dilution reduced the temperature gradients produced in the mixing region and maintained an overall temperature compatible with an experimental rig constructed of conventional materials.

The use of this particular chemical system (CO-O₂-H₂O-N₂) offers two advantages over other possible chemical systems. The first is that preheating the reactants makes them mutually self-igniting and no piloting is necessary to provide a heat source to sustain the reactions. Consequently, reactant concentrations outside the ordinary flammability limits may be used to minimize the overall heat release and the effect of chemical reaction on the flow field. The second advantage is that the molecular weight of the components do not vary greatly. The combined effect of these two features is that problems associated with the sampling of gases in flows with large density gradients (ref. 9) (i.e., hydrogen diffusion flames) are avoided.

Experimental Hardware

The experimental study was carried out in a confined coaxial flow test rig shown in figure 1. The apparatus was fabricated from Type 304 stainless steel and was insulated to minimize heat losses.

Hydrogen was injected into the air stream through three hundred and sixty 0.078 centimeter holes in six 0.317 centimeter stainless steel tubes. The hydrogen burned as diffusion flames.

The heat exchanger was used to preheat the center stream by heat transferred from the hot outer stream. It consisted of four U tubes manifolded into the center pipe. The tubes were fabricated from 1.58 centimeter I.D. pipe. The overall tube length was 205 centimeters.

The test section as shown in figure 2, consisted of an outer pipe with a 10.2 centimeter inside diameter and a center pipe with a 3.34 centimeter outside diameter (2.66 cm I.D.) tapered to a 0.05 centimeter lip to minimize wake effects.
Turbulence generators (perforated plates and grids) were placed in the annular flow passage, 21.6 centimeters upstream of the exit plane of the center pipe. The perforated plates shown in figure 3 were 0.32 centimeter thick, Type 601 Inconel disks with 0.635 centimeter holes. The grids shown in figures 4-5 were formed from 0.64 and 0.32 centimeter Type 304 stainless steel rods. Blockages of 68% and 47% were used for each set of turbulence generators.

The mesh length for the grids was taken as the distance between the centers of the grid elements as shown in figure 6. For the large rod grids, this distance was 1.27 centimeters and for the smaller rod grids, 0.635 centimeters.

For the perforated plates, the mesh length was defined by the size of a square of material surrounding the hole as shown in figure 6. The hole distribution was not uniform for the perforated plates used in this study. Hence the mesh length for these plates was calculated for a fictitious square plate of solidity and hole size equal to the actual perforated plates. The equivalent mesh length was 0.99 centimeter for the 68% blockage plate and .77 centimeters for the 47% blockage plate.

**Species Measurements**

Measurements of carbon monoxide and carbon dioxide mole fraction were obtained with a continuous flow gas sampling and on line gas analysis system. The gas samples were drawn at various locations. Axial positions varied from the center pipe exit plane to 23 centimeters downstream. Radial locations varied from the pipe centerline to the outer pipe wall.

A water cooled, supersonic, gas sample probe was used to withdraw gas samples. Previous research (ref. 10) has shown that this type of probe is capable of effectively quenching the reactions in the CO-O2-H2O chemical system. While this type of probe does not yield fluctuating composition measurements, it does allow the measurement of the overall extent of reaction due to mixing at a point in the flow. It is also much simpler to operate than measurement systems which do yield time resolved, fluctuating composition data.

The probe, shown in figure 7, was fabricated from type 304 stainless steel tubing arranged concentrically to form flow passages for the gas sample and cooling water. The combustion gases were passed through a sonic orifice at the tip of the probe, expanded through a 16:1 area ratio nozzle and flowed through the center tube to the exit at the rear of the probe.

The sample collected by the probe was transported to the analysis instruments through stainless steel tubing in order to prevent any further chemical reactions.
The specie mole fractions of carbon dioxide and carbon monoxide were measured simultaneously using two nondispersive infrared analyzers (Beckman Instruments Model 315B instrument accuracy 1% full scale). A dry-ice-cooled water trap was utilized to prevent water vapor from condensing in the sample lines or analyzers and interfering with the measurements. It was necessary to pump the gas sample to achieve supersonic flow in the probe since the tests were conducted at nearly atmospheric pressure.

Velocity and Turbulence Measurements

Measurements of the mean velocity, the axial turbulent intensity, the integral and dissipation scales were performed. A constant temperature anemometer system was utilized in conjunction with several signal analysis instruments. A cylindrical hot film sensor was used. Measurements were performed at 0.15 centimeters downstream of the center jet exit plane. The sensor design established this as the closest measurement location.

Test Conditions

The experiments of this research were carried out in two phases. The specie measurements were performed in high temperature combustion tests while the turbulence measurements were obtained in ambient temperature tests. The flow rates for these ambient temperature tests were selected to simulate the turbulent characteristics of the high temperature flow and was done on the basis of Reynolds number similarity.

The experiments were conducted near atmospheric pressure with the streams at equal bulk velocities and temperatures. The test conditions for the majority of the experiments are summarized in Tables 1 and 2. As will be discussed later, several experiments were also conducted with the initial carbon monoxide concentrations reduced from 9\% to 2\%. In addition, preliminary experiments were conducted over a range of initial temperatures from 1000 K to 1300 K.

RESULTS AND DISCUSSION

An experimental study of turbulent reactive mixing was performed to investigate the effects of altering turbulent mixing characteristics upon the extent of chemical conversion. The mixing characteristics were altered by placing turbulence generators in the flow field.

Ambient Temperature Experiments

For each of the turbulence generators and for the no blockage case (no turbulence generator), traverses with a hot film sensor were performed to obtain the turbulent flow characteristics. The center stream was the same for all cases.
Mean velocity and turbulent intensity profiles. - The data shown in figure 8a-c reveal that the mean velocity profile of the center stream \((r/Ro < 1)\) has not yet evolved into the profile which is associated with fully developed pipe flow. This was anticipated since the center pipe has a length to diameter ratio of approximately 20 and the establishment of fully developed flow requires at least 40 tube diameters.

The curves on each of figures 8a-c are for the no blockage condition. These curves show the velocity profile in the annulus \((r/Ro > 1)\) is relatively flat. The turbulence generators produced an unexpected effect in that they altered the annular mean velocity profile. The 68% blockage plate and the 47% grid with 0.64 centimeter rods flattened the profile while the other generators skewed the maximum velocity point toward the center pipe wall.

The turbulent intensity of the center stream \((r/Ro < 1)\) was 0.08 on the centerline and is twice the value reported for fully developed flow in a smooth pipe (ref. 11). This high value probably results from the opposing flow configuration of the inlet flows into the center pipe from the four heat exchanger tubes.

The turbulent intensity of the no blockage annular flow \((r/Ro > 1)\) is approximately 0.08 and probably was a result of the presence of the heat exchanger in the upstream section (figure 1). As shown in Table 3, three of the turbulence generators increased the turbulent intensity while the other three reduced it. This behavior is a result of the initial level of turbulence generated by the device and its decay downstream. The higher blockage devices act to increase the initial level of turbulence. The differences between the turbulence generated by devices of equivalent blockage are related to the decay of the turbulence; the smaller scale turbulence generated by the smaller grids decays more rapidly.

Scales of turbulence. - The measurements of the dissipation scale and integral scale were performed at the maximum velocity point. The results are summarized in Table 3. The dissipation scales are roughly equivalent, which would be expected as the same fluid conditions were used and hence the same viscous forces were acting.

The variations in the integral scales can roughly be related to the geometrical characteristics of the turbulence generators. This was done by nondimensionalizing the integral scale and the distance downstream of the turbulence generator with respect to the turbulence generator mesh lengths.

The normalized integral scales as a function of the number of mesh lengths downstream of the turbulence generators are shown in figure 9. The growth of the normalized integral scale is consistent with observations of other experimenters (refs. 12-13).
Combustion Experiments

Effects of temperature. - Since very high temperatures significantly affect the useful lifetime of experimental apparatus, it was desired to operate the test rig at a minimum temperature above which no change in the composition measurements were observed.

The operating temperature was established by a series of preliminary experiments in which flow field surveys of the gas composition were performed at various temperatures. Figure 10 shows the effect of temperature upon the extent of reaction at a point in the flow and indicates the selected operating temperature of 1280 K to be satisfactory since no composition changes occur above this temperature.

Effect of initial carbon monoxide concentration. - The effect of the initial amount of carbon monoxide in the center jet was investigated. In these experiments, the carbon monoxide concentration was reduced to 2% and the results were compared to surveys with an initial carbon monoxide level of 9%.

It was found that within experimental error, no variation of the mass fraction profiles (fraction of sample gases which originated from the center stream) or the extent of conversion profiles could be discerned. This indicates that:

1. the chemical reaction did not significantly affect the flow field and
2. that the rate of mixing on the molecular level was not dependent upon the initial composition of the flow.

Specie profiles. - Measurements of carbon monoxide and carbon dioxide were obtained using the sampling and analysis system previously described. From these measurements and the calculated initial compositions of the streams, a complete determination of the wet basis specie mole fractions was obtained. Radial specie profiles for the 47% blockage plate are shown in figures 11a-f, at increasing distances downstream of the exit plane. The specie mole fractions are nondimensionalized with respect to the initial amount of carbon monoxide in the center stream. The nondimensional value of oxygen in the annular flow exceeds unity because the oxygen mole fraction in the annular flow is greater than the initial amount of carbon monoxide.

These figures which are for the 47% blockage plate show the general behavior observed in all the cases investigated, (ref. 14). The carbon monoxide profile (figure 11a) is discontinuous at the center pipe exit plane, having a constant value of unity across the center jet and dropping to zero
at the edge of the jet. The value of carbon monoxide on the center line will remain constant until the end of the potential core of concentration is reached. It then decreases as carbon dioxide and oxygen are transported into the downstream center regions. Eventually, the carbon monoxide fraction goes to zero as the oxidation process is completed. From this point onward, the mixing is unreactive as the carbon dioxide and excess of the outer stream are distributed to a final uniform mixture. The composition of this mixture will depend upon the relative initial mass flows of the streams and their compositions.

In these pseudo-hypergolic mixing experiments any mixing of carbon monoxide and oxygen at the molecular level will result in carbon dioxide formation. The reaction time is approximately 130 μseconds. The measurement of carbon monoxide-oxygen coexistence in certain regions results from the turbulent mixing process. The gas composition ingested by the sample probe fluctuates as eddies containing only CO2/N2 or O2/N2/H2O, separated by regions of CO2/N2/H2O, are swept by the sampling location. The sample gases contain both carbon monoxide and oxygen because the reactants had not mixed on a molecular level even though bulk mixing of the reactant eddies had occurred.

Mass fraction of center stream. - Radial profiles of the mass fraction of center stream (the fraction of the sample gases which originated from the center stream) are presented individually in figures 12a-g. Mass conservation was checked at each axial sampling location. Deviations up to ± 10% of the calculated values were found. The data are included for completeness and are typical of coaxial flow mixing.

Under the influences of the turbulent bulk mixing processes, the center stream spreads as eddies transport mass into the outer annular regions. Downstream of the potential cores, this results in a decay of the centerline mass fractions. These results are summarized in figure 13. The slope of the decay rate for the various turbulence generators lie in the -1.0 to -2.0 range. These are well within the bounds of -0.5 to -3.0 which has been reported by Harsha (ref. 15) for coaxial jet mixing. In this study, the lengths of the potential cores are much shorter than those reported by Harsha. This is probably due to the high initial turbulent intensities of the flows. The experiments reported by Harsha utilized flows with low initial intensity.

Extent of reaction. - The individual radial profiles of the extent of carbon monoxide conversion are shown in figures 14a-g. As the reactants are mixed by the turbulent flow field, the fraction of carbon monoxide converted to carbon dioxide increases. The values asymptote to complete conversion at the edge of the center jet and will drop to zero beyond the edge of the jet since no carbon monoxide is available for conversion. The extent of conversion is limited by the degree of molecular level mixing and not chemistry.
Comparison of turbulence generators. - Turbulence acts to break up reactant streams to allow increased surface area for the interdiffusion of the reactants. Since reactant conversion is determined by this interdiffusion, any process which leads to an increase in the contact area available for intimate mixing will also increase the conversion. Increased conversion may be brought about by various means:

1. Increase the bulk mixing to increase the number of reactant clumps in contact while holding their size fixed;

2. Decrease the eddy size while holding the bulk mixing constant; or,

3. Combining these mechanisms to increase bulk mixing and reduce the average eddy size.

These features may be observed by comparison of the centerline data for mass fraction (figure 13) and conversion (figure 15) and the average eddy size, Table 3. In particular, the centerline mass fraction of the 68% blockage, 0.64 centimeter grid follows that of the no blockage case indicating equivalent bulk mixing. However, the size of the eddies is smaller in the no blockage case and hence, there is an increased amount of conversion for any particular level of bulk mixing. The centerline mass fraction of the 47% blockage plate lies below the levels of the no blockage case which indicates more bulk mixing. However, the scale of the former are larger than the no blockage case with the net result being nearly equivalent levels of conversion.

The relatively poor conversion of the 0.32 centimeter rod grids is seen to lie in the large eddy which they produce rather than poor bulk mixing.

The 68% blockage plate yields the best results of the turbulence generators tested because it has the best bulk mixing and produces a comparatively small scale.

These centerline plots illustrate that complete reactant conversion does not necessarily imply complete mixing at the molecular level. By virtue of the relative mass flows of the two streams, a completely uniform mixture would have a center stream mass fraction of approximately 0.13. Extrapolation of the conversion and mass fraction plots reveal the carbon monoxide to be completely converted before the mass fractions reach this level. Thus, the center stream has been sufficiently molecularly mixed to accomplish the complete conversion and the relaxation to complete uniformity will involve the unreactive mixing of the carbon dioxide and the excess oxygen stream.

The centerline plots also illustrate the difference between unreactive and reactive mixing. Mass fraction decay plots are typically used to characterize unreactive coaxial mixing and on this basis one would consider the no blockage case to be one of the more poorly mixed since less of the outer stream is present in the sample gases than in other cases. However, in terms
of conversion, it is among the best. Thus, unreactive flow computations
do not reveal the true picture of reactive flows.

SUMMARY

A continuous flow, gaseous, pseudo-hypersonic mixing experiment has been
developed to investigate the mixing characteristics of turbulent flow fields.
A carbon monoxide-oxygen-water vapor chemical system was selected on the
basis of facilities, safety, and compatibility with the use of a continuous
gas sampling and analysis system. The reactant streams were preheated to
1280 K to approach infinitely fast chemical reactions.

Experiments were performed in a coaxial mixing configuration. Six tur-
bulence generators were used to alter the mixing characteristics of the
reacting flow.

A water cooled, sonic orifice gas sampling probe was utilized to obtain
gas composition measurements.

In the course of the investigations, it was found that:

1. The six-turbulence generators altered the turbulent mixing field
which affected the degree of conversion of the carbon monoxide
and the mass fraction of the center stream.

2. Increasing the bulk mixing and decreasing the average eddy size
resulted in a higher degree of conversion.

3. The degree of molecular mixedness was not dependent upon the
initial reactant concentrations.

4. The heat release due to carbon monoxide oxidation did not signifi-
cantly affect the flow field.

5. Increasing the reactant temperatures above 1280 K did not change
the amount of carbon monoxide conversion for a given geometry.
REFERENCES


### TABLE 1. COMBUSTION EXPERIMENTS - TEST CONDITIONS

<table>
<thead>
<tr>
<th>Specie</th>
<th>Flow rate, kg/sec</th>
<th>Temperature, K</th>
<th>Velocity, m/sec</th>
<th>Reynolds number</th>
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<tbody>
<tr>
<td>Center stream</td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>N₂</td>
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<td>1280</td>
<td>50</td>
<td>7600</td>
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<tr>
<td>CO</td>
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<tr>
<td>Annular stream</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Air</td>
<td>63.0x10⁻³</td>
<td>1280</td>
<td>50</td>
<td>34000</td>
</tr>
<tr>
<td>H₂</td>
<td>1.1x10⁻³</td>
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### TABLE 2. AMBIENT TEMPERATURE EXPERIMENTS - TEST CONDITIONS

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<th>Temperature, K</th>
<th>Velocity, m/sec</th>
<th>Reynolds number</th>
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<tbody>
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<td></td>
</tr>
<tr>
<td>N₂</td>
<td>2.95x10⁻³</td>
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<td>3.7</td>
<td>7600</td>
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<td>Annular stream</td>
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<td></td>
</tr>
<tr>
<td>Air</td>
<td>3.9x10⁻²</td>
<td>280</td>
<td>3.7</td>
<td>34000</td>
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### TABLE 3. TURBULENCE CHARACTERISTICS 21.6 CM DOWNSTREAM OF TURBULENCE GENERATORS

<table>
<thead>
<tr>
<th>Turbulence generator</th>
<th>Minimum turbulent intensity</th>
<th>Integral scale, cm</th>
<th>Dissipation scale, cm</th>
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<td>47% Blockage plate (0.635 cm holes)</td>
<td>0.09</td>
<td>1.15</td>
<td>0.04</td>
</tr>
<tr>
<td>68% Blockage plate (0.635 cm holes)</td>
<td>0.09</td>
<td>0.98</td>
<td>0.05</td>
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<tr>
<td>47% Blockage grid (0.64 cm rods)</td>
<td>0.065</td>
<td>1.15</td>
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<td>68% Blockage grid (0.64 cm rods)</td>
<td>0.092</td>
<td>1.17</td>
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<td>47% Blockage grid (0.32 cm rods)</td>
<td>0.065</td>
<td>1.4</td>
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<td>68% Blockage grid (0.32 cm rods)</td>
<td>0.075</td>
<td>1.2</td>
<td>0.039</td>
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<tr>
<td>No blockage</td>
<td>0.082</td>
<td>0.95</td>
<td>0.04</td>
</tr>
<tr>
<td>Center stream (all cases)</td>
<td>0.08</td>
<td>1.07</td>
<td>0.04</td>
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*REPRODUCIBILITY OF THE ORIGINAL PAGE IS POOR*
Figure 1. Confined coaxial flow apparatus.

Figure 2. Coaxial flow test section.
Figure 3. - Inconel, 10.2 cm diameter perforated plate turbulence generators with 0.635 cm holes.

Figure 4. - Grid turbulence generators of 0.64 cm, type 304 stainless steel rods.

Figure 5. - Grid turbulence generators of 0.32 cm, type 304 stainless steel rods.
Figure 6. - Definition of mesh length (M) for grids and perforated plates.

Figure 7. - Internal schematic - gas sample probe.
Figure 3. - Mean velocity and turbulence intensity at jet exit plane.

(a) BLOCKAGE PLATES, 0.635 cm DIAMETER HOLES.

(b) BLOCKAGE GRID, 0.64 cm DIAMETER RODS.
Figure 8. Concluded.
Figure 11. 47% Blockage plate, radial distribution of normalized species.
Figure 11. - Continued.

(c) $X/D_o = 2.86$.

(d) $X/D_o = 4.76$.

Figure 11. - Continued.
Figure 11. - Concluded.
Figure 12. - Radial distributions, mass fraction of gases originating from center stream.
Figure 12. - Continued.
Figure 12. - Continued.

(e) 68% BLOCKAGE GRID (0.32 cm RODS).

(f) 47% BLOCKAGE GRID (0.32 cm RODS).

RADIUS RADIUS, r/R_0
Figure 12. - Concluded.

Figure 13. - Centerline values, mass fraction of gases originating from center stream.
Figure 14. - Radial distributions, fractional conversion of CO.
Figure 14. - Continued.
Figure 14. - Continued.
Figure 14 - Concluded. 

Figure 15 - Centerline values, fractional conversion of CO to CO$_2$. 

(g) NO BLOCKAGE CASE.