

**Investigation of NO_x and CO Formation in Lean-Premixed, Methane-Air,
High-Intensity, Confined Flames at Elevated Pressures**

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Abstract

The coupling between NO_x formation chemistry and the mixing/transport environment is of critical importance to the design of lean-premixed gas turbine combustors, but is incompletely understood. In the present research, this problem is addressed via the study of NO_x formation in a high-pressure jet-stirred reactor operating on lean-premixed methane/air. These experiments focus on the effects of residence time (0.5-4.0 ms), pressure (3.0, 4.7, and 6.5 atm), and inlet temperature (344-573 K). The combustion temperature varies from 1815±5 K at the lowest residence times to 1910±30 K at the largest residence times. The NO_x is lowest at intermediate residence times, reaching higher values at the extremes. Increasing pressure and inlet temperature tend to reduce NO_x concentrations. Concentration profiling in the reactor suggests two general environments: (1) a highly non-equilibrium reaction zone defined by high CO concentrations, and (2) a post-flame environment. The NO_x formation is concentrated in the region of strongly non-equilibrium combustion chemistry. The Damköhler number is $0.06 \leq Da \leq 1$, and the ratio of turbulent intensity to laminar burning velocity is $28 \leq u'/S_L \leq 356$, indicating the combustion occurs in the high intensity, chemical rate limiting regime. The results are interpreted using a two-environment, detailed chemistry model in which the size and structure of the flame environment are established by matching the measured data, and which is independently verified using turbulent flame velocity/thickness correlations. The modeling suggests NO_x formation is controlled by both the specific conditions in the non-equilibrium zone and by the size of the zone. Since both these features are influenced by the experimental parameters, a highly non-linear scenario emerges with implications for minimizing NO_x via combustor design. The modeling also suggests the unique case of well-stirred combustion for NO_x at elevated pressure is obtained at low residence time conditions.

Introduction

Lean-premixed gas turbine combustors provide an economical means of achieving very low NO_x emissions. The design of these combustors is a challenging problem, involving the use of complex configurations and flow fields to achieve a compromise between low NO_x performance and acceptable flame stability. Tests conducted on a number of prototype and commercial designs show the variation of NO_x with common combustion parameters is far from consistent [1]. In some designs high pressure leads to markedly higher NO_x, while in others the NO_x decreases with pressure. Measurements suggest some of the complexity occurs because the external parameters change the size of the zones where NO_x forms, in addition to changing the rate of formation within the zones. The purpose of the present work is to explore how this behavior occurs in lean-premixed, high-intensity combustion, such that insight can be gained for the combustor design problem.

The high-pressure jet-stirred reactor (HP-JSR) has been used before as an experimental idealization of lean-premixed gas turbine combustion. Steele [1] employed a HP-JSR geometrically similar to the present reactor to determine the response of NO_x to combustion temperature for lean-premixed methane combustion. The activation temperature for the NO_x (adjusted to 15% O₂ dry) was found to be 23650 K. Additionally, the NO_x was observed to decrease with increasing pressure (1-7.1 atm), and to be insensitive to inlet temperature (to 600K) and residence time (2-4 ms). Bengtsson et al. [2] examined the effect of pressure (1-20 atm) at a fixed methane-air stoichiometry ($\phi=0.55$), with the residence time varied from 1 to 2 ms in a multi-nozzle jet-stirred reactor. The measurements show a decrease in NO_x with pressure, with the effect becoming negligible above 6 atm.

In the present work, the focus is on the influence of residence time, which is varied from 0.5 to 4.0 ms. Also, the effects of pressure and inlet temperature on NO_x are examined over this range. Previous work [1] recognized the HP-JSR is not spatially uniform at high pressure, so the present work focuses on resolving the structure of the reacting gas, on interpreting the results via modeling of the structure, and on seeking conditions for which the HP-JSR operates well-stirred for NO_x formation.

Experimental Setup and Conditions

The high-pressure experimental rig, with the HP-JSR, is shown in Fig. 1. Details are given in [1]. Prior to entering the HP-JSR, the combustion air passes through the air preheater (electric) and premixer, all located inside the pressure vessel. A second air stream that bypasses the HP-JSR is used to regulate the pressure within the vessel. The volume of the HP-JSR, i.e., the axisymmetric jet-stirred cavity, is 1.5 cm³, with dimensions as shown in the Fig. 1. The reactor material is cast alumina. Because of the long residence time in the premixer (100 ms) and the mixing within the premixer and inlet jet, the HP-JSR is assumed to operate fully premixed.

The HP-JSR is run over the 0.5 to 4.0 ms residence time (τ) range. Because of the almost eight-fold range in flow rate, the jet velocity and pressure drop across the nozzle vary significantly. For operation at 0.5 ms residence time, the pressure drop is 45% and the jet inlet velocity is close to sonic. Whereas, for operation at 4.0 ms residence time, the pressure drop is about 3% and the jet inlet velocity (without preheat) is 75 m/s.

Species and temperature profiles are obtained along the path (at two-thirds reactor height) indicated in Fig. 1. Gas sampling is done with a small ceramic probe. Analyzers measure NO, NO_x, CO, CO₂, and O₂. Because the majority of NO_x found in the sample line is NO₂, and NO₂

is soluble in water, the sample line is maintained above the condensation temperature upstream of the ice bath condenser located just upstream of the analyzers. Limited testing with a NO_x-to-NO converter placed in the sampling line is used to confirm negligible loss of NO₂ in the condenser.

The gas temperature measurements are obtained with R-type thermocouples, coated with Ceramabond 569 (a high temperature paste containing $\geq 99\%$ alumina). The thermocouple is placed in the middle of the recirculation zone, on the side of the reactor opposite to that containing the sampling probe. The recirculation zone represents the bulk of the reactor, and is nearly uniform in temperature and species concentrations. The thermocouple temperature is maintained at 1803 ± 5 K by tuning the fuel-air equivalence ratio (ϕ), and is corrected to obtain the true gas temperature (termed the combustion temperature) using two methods as described in [3]. In the first method, the convective gain and radiative loss of the thermocouple are analyzed. In the second method, the heat loss of the HP-JSR is calculated. The two methods give similar values, which are averaged to obtain the combustion temperature. For residence times below about 1.0 ms, the convective loading onto the thermocouple is large, and thus the thermocouple temperature requires little correction. Furthermore, the HP-JSR is nearly adiabatic. For larger residence times, the convective loading onto the thermocouple weakens, hence the combustion temperature is greater than the thermocouple temperature. Additionally, the reactor is no longer adiabatic. The result is a combustion temperature of 1815 ± 5 K at the lowest residence times. For the experiments at 4.7 and 6.5 atm, the combustion temperature increases to about 1880 K at the largest residence times, whereas for 3.0 atm, it reaches about 1940 K at the largest residence times. Mean ϕ is 0.71 for the experiments without inlet preheat, and 0.62 with preheat. The range is $0.58 \leq \phi \leq 0.80$. Details on the variation of ϕ with reactor conditions are given in [3].

Experimental Results and Discussion

The effect of residence time on measured NO_x is shown in Fig. 2, for pressures of 6.5, 4.7 and 3.0 atm, without preheating of the reactants. The measurements are obtained 3 mm from the reactor axis, in the middle of the recirculation zone.

Starting at the lowest residence times, the NO_x is seen to decrease with increasing residence time, reach a minimum, and then increase. The increase is caused by the increase in the combustion temperature for the largest residence times.

The NO_x data for the 4.7 and 6.5 atm experiments can be compared for the effect of pressure. For identical residence time, the experiments have nearly identical combustion temperature. Thus, the increase in NO_x with decreasing pressure noted in Fig. 2 is not masked by changing temperature. For the 3.0 atm experiments, the combustion temperature at residence times larger than 1.5 ms is greater than the temperature for the 4.7 and 6.5 atm experiments. Thus, the significant increase in NO_x for the 3.0 atm data at the intermediate and large residence times is caused at least in part by the temperature increase.

Figure 3 shows the NO_x results for the reactor inlet gas preheated to 573 K. Because of the preheat, the reactor operates leaner than the unheated inlet experiments. As above (in Fig. 2), the NO_x increases as the pressure decreases from 6.5 to 4.7 atm. Also as above, the increase in NO_x effected by decreasing the pressure to 3.0 atm is influenced by the greater combustion temperature of the 3.0 atm measurements. Comparison of Fig. 3 with Fig. 2 indicates less NO_x formation with the preheated inlet. The effect is most pronounced at the low residence times, where the combustion gas temperature is about 1815 K for all cases. For 1.0 ms, using the best

curve fit results, the reductions in NO_x affected by preheating the inlet jet are about 20% at 3.0 atm and about 30% at 6.5 atm.

Adjustment of the NO_x data to the reference condition of 15% O₂ dry, followed by an additional adjustment to the average temperature (1815 K) of the low residence time experiments, using the activation temperature of 23650 K of Steele [1], shows a decrease in NO_x as the residence time increases from 0.5 ms up to 3.0 ms. For residence times greater than 3.0 ms, the adjusted data increase slightly. For the unheated inlet experiments, the adjusted NO_x (based on best curve fits) decreases from 4.5±0.2 ppmv at 1.0 ms to 3.2±0.3 ppmv at 3.0 ms, whereas for the heated inlet, the adjusted NO_x changes from 4.2±0.2 ppmv at 1.0 ms to 3.4±0.2 ppmv at 3.0 ms. The upper limits in the NO_x uncertainties pertain to 3.0 atm pressure, the lower limits to 6.5 atm. The adjusted results indicate there is a minimum value of NO_x of about 3.0 ppmv (15% O₂ dry) for combustion at 1815 K. Additionally, the adjustment essentially eliminates the reduction of NO_x with inlet preheat noted above.

Examples of spatial profiles of CO and NO_x at 6.5 atm are shown in Figs. 4 and 5. The CO is low on centerline, since this represents fresh, largely unreacted gas, and reaches a peak in the shear region at the edge of the jet, indicating this to be the location of intense reaction. As noted in Fig. 1, the jet impinges on the top wall of the reactor and recirculates down the inside wall of the reactor. For large residence times, CO has ample opportunity to complete reaction, and the recirculation zone shows a flat profile of relatively low CO concentration. At the low residence times, the time is insufficient for CO to complete burnout, as evidenced by elevated CO in the recirculated gas. For 0.5 ms, except for the center of the recirculation zone, the CO is approaching a level of about 1%. For the cases involving preheat (only 0.9 ms shown), the CO is lower due to leaner stoichiometry. The NO_x in Fig. 5 exhibits nearly uniform concentration in

the recirculation zone. That is, the recirculation zone appears well-stirred for NO_x. This can happen if NO_x enters the recirculation zone and is well dispersed. It can also happen if NO_x forms in the recirculation zone at a rate slow compared to the mixing rate. Because of the NO_x uniformity, and because the exhaust is drawn from the recirculation zone, the recirculation zone NO_x concentration can be regarded as the exhaust NO_x concentration of the reactor.

Modeling and Discussion

The physical picture that emerges of the situation just described is of a turbulent premixed flame anchored over and around the inlet jet, followed by a post-flame environment. Because the post-flame environment is stirred, the CO and free radical species do not relax to equilibrium as quickly as for a plug flow reactor. The size of the turbulent flame (*i.e.*, the fraction of the reactor volume occupied by the highly non-equilibrium gas) increases for low residence times. At the lowest times, it may fill the entire reactor. This situation lends itself to a two environment chemical kinetic model in which the first zone represents the non-equilibrium turbulent flame and the second the post-flame gas. The partitioning into the two environments depends on the turbulent premixed flame regime present in the reactor.

The Damköhler number, $Da = \text{mixing time}/\text{chemical time}$, and the turbulence Reynolds number, $Re_T = u' d_o / \nu$, are estimated for the flames in the present work. The mixing time is calculated as d_o / u' , where d_o is the jet nozzle diameter, which is assumed to be the turbulence integral scale (l_o), and u' is the turbulent intensity, which is assumed to be 10% of inlet jet (cold) velocity (u_o). Chemical time (τ_L) is calculated as α / S_L^2 , where α is the thermal diffusivity and S_L is the laminar burning velocity, calculated by equations given by [4]. The kinematic viscosity (ν) and α are determined for the inlet temperature and reactor pressure.

The $Da-Re_T$ range for the present experiments is $Da \cong 0.06/Re_T \cong 10^4$, corresponding to the lowest residence times, to $Da \cong 1.0/Re_T \cong 10^3$, corresponding to the largest residence times with the reactor operating at the highest temperature levels. The $Da-Re_T$ range indicates the operation of the HP-JSR falls in the flamelets-in-eddies regime [5]. This regime is characterized by torn flamelets that can be non-contiguous, *i.e.*, a thick turbulent reaction zone containing "islands" of reactants and products in which the reactant pockets are strained by the flow while the reaction front moves into their periphery.

The turbulent flame thickness is used to evaluate the fraction of the reactor occupied by the non-equilibrium flame zone. This turbulent flame thickness, δ_T , is determined by analogy to the laminar flame thickness as the ratio of the turbulent diffusivity ($D_T \cong u' l_o$, [6]) to the turbulent burning velocity, S_T , *i.e.*, $\delta_T \cong u' d_o / S_T$. Velocity S_T is calculated based on the equation suggested by [7]: $S_T = S_L + u' (1+Da^{-2})^{-1/4}$. For small Da and $u' d_o / \delta_L \gg S_L$, which is the case in the present work, the S_T equation reduces to: $S_T \cong u' Da^{0.5} \cong (u' d_o / \tau_L)^{0.5}$, which is the turbulent analogy to the laminar burning velocity. Consequently, the flame thickness formula reduces to $\delta_T \cong d_o / Da^{0.5}$. The resulting values for δ_T vary between 1.6 mm (for large residence time) and 5.4 mm (for small residence time). Thus, in some cases the flame may fill the reactor volume since the reactor radius (at its widest point) is 5.75 mm.

The low Damköhler number and high turbulence intensity suggest the flame zone may be modeled to first approximation as a perfectly-stirred reactor (PSR). The two environment model thus becomes two PSRs in series with the second representing the stirred post-flame environment. Earlier applications of modeling with two PSRs are found in [1] and [2]. The only parameter requiring selection is the fraction of the reactor volume associated with the first PSR (PSR1). One approach is to select this based on δ_T . An independent approach is to determine

the volume of PSR1 by matching the measured NO_x and CO to values calculated by the two PSR modeling. The result derived can then be compared against the flame volume inferred from δ_T .

Matching of the model to the measured NO_x and CO is performed by varying the volume of PSR1 from the blowout volume, determined based on the prescribed fuel-air equivalence ratio and reactor mass flow rate, to the full reactor volume (1.5 cm³). The chemical kinetic mechanism used in the two PSR calculations is GRI-Mech. 3.0 [8].

Figure 6 shows how the calculated NO_x and CO concentrations vary with the fraction of the total reactor volume assigned to PSR1. The residence times are 0.5 and 4.0 ms, the pressure is 6.5 atm, and the inlet jet is not preheated. Focusing on the 0.5 ms case, the CO is relatively constant for all PSR1 volumes until the size of PSR2 (which acts as the relaxation zone) becomes quite small. The NO_x remains constant unless the size of PSR1 approaches blowout. For this case, the data are CO: 4110 ppmv, NO_x: 12 ppmv. The NO_x can thus be matched by a large range in PSR1 volumes, but the CO can be matched only when PSR1 approaches 100% of the reactor volume. For this condition, the two PSR model can match the data only by assuming the non-equilibrium flame zone essentially fills the reactor, which is consistent with the experiments at low residence times. For the 4.0 ms case, the data are: CO=190 ppmv, NO_x=10.3 ppmv. This CO level is matched by most PSR1 volumes, indicating CO is near equilibrium. Only a very small PSR1 can match the NO_x data. This implies a situation where the flame zone is small and the bulk of the reactor can be treated as a post-flame relaxation zone, again consistent with the experiments.

The NO_x formation rate is significantly higher in PSR1 for all cases, due to the higher O-atom and CH concentrations promoted by the fuel oxidation. For the 0.5 ms case in Fig. 6, the

O-atom concentration increases quickly as the volume of PSR1 increases from the blowout condition. This accounts for the rapid increase in NOx formation as the conditions step away from blowout. At longer times, the O-atom decreases with increasing PSR1 volume, but the increased time available for NOx formation essentially counterbalances the effect, leaving NOx nearly constant with residence time. Another contributing factor is NOx formation by CH+N₂. The CH concentration mirrors the trends of the O-atom, but is more extreme in its fall-off with increasing residence time, decreasing by over a factor ten as the PSR1 volume increases over its full range. Thus, the NOx formation chemistry is influenced by both the size of the non-equilibrium zone and by the conditions within the zone.

The volume partition resulting from the two PSR modeling (via matching CO and NOx) is shown in Fig. 7 for 6.5 and 3.0 atm without preheat. For low residence times, data are matched for volume ratios of about 100% (i.e., the flame fills the reactor), whereas at large residence times the matching occurs closer to a few percent of reactor volume (i.e., a small turbulent flame).

Also shown in Fig. 7 is the percentage of the reactor occupied by the turbulent flame, calculated from the turbulent flame modeling. With the half-angle of the unreacted jet cone taken as $\theta \cong d_o/2h \cong S_T/10 u'$, where h is the cone height and the inlet velocity is $u_o = 10u'$, one obtains $h \cong 5\delta_T$. The corresponding flame volume is $V_{\text{flame}} \cong 5\pi\delta_T^3$. This is used to calculate the V_{flame}/V_t values plotted in Fig. 7; V_t is the reactor volume. When h becomes approximately equal to the height of the reactor cavity ($L = 20$ mm), $\delta_T \cong 3.6$ mm and $V_{\text{flame}} \cong 0.75$ cm³. This is one-half the reactor volume. Comparison to the two PSR modeling shows this condition closely corresponds to the sharp transition from $V_{\text{psr1}}/V_t = 100\%$ to $V_{\text{psr1}}/V_t < 10\%$. The Damköhler number at transition is $Da \cong 0.15$.

As noted in Fig. 7, the 3.0 atm case undergoes transition at a smaller residence time than the 6.5 atm case. Recall that the 3.0 atm measurements have a higher combustion temperature than the 6.5 atm measurements at intermediate and large residence times. For these residence times, the 3.0 atm flame has a greater Da than the 6.5 atm flame, and hence, a smaller thickness. In order for the 3.0 atm flame to fill the reactor, the reactor must operate at a lower residence time than for the 6.5 atm case. Also, the 3.0 atm case shows closer agreement between the flame thickness modeling and the two PSR modeling, probably because its temperature and hence Da change significantly between low residence time (1815 K) and large residence time (1940 K).

Chemical Kinetic Discussion

Results from the chemical kinetic modeling of the unheated inlet experiments (all pressures) and the heated inlet experiment at 6.5 atm indicate the following when the reactor is operating in single PSR mode (residence time $0.5 \leq \tau \leq 1.5$ ms):

- [O], [H], and [NNH] decrease 2-to-3-fold over the 3-fold increase in τ , [N₂O] is nearly constant, and [CH] decreases by 8-fold. Temperature is nearly constant. The nitrous oxide (N₂O+O,H) and prompt (CH+N₂) reactions are predominant for NO formation. By the nitrous oxide mechanism, the NO yield is relatively insensitive to τ , because the increase in τ is offset by the decrease in [O] and [H], and by the prompt mechanism, the NO decreases because of the sharp decrease in [CH]. The fall-off in measured NO_x with increasing τ (Fig. 2) for the low residence times is consistent with the decrease in [CH].
- [O] is relatively insensitive to preheat, while [H] and [NNH] decrease slightly and [N₂O] increases slightly. [CH] decreases 5-fold. Thus, the decrease in NO_x obtained by

preheating the experiment and the flatness of the NO_x data in Fig. 3 for 6.5 atm ($\tau \leq 1.5$ ms) are consistent with the diminished influence of [CH].

- [O], [H], and [NNH] increase with decreasing pressure, while [N₂O] decreases almost 2-fold, and [CH] is insensitive. Overall, the NO yield (both modeled and measured) increases with decreasing pressure.

For the two PSR regime ($\tau \geq 1.5$ ms), 74 ± 10 % of the modeled NO_x forms in PSR1 when $\tau = 2$ ms, whereas for 4 ms, the value is 54 ± 7 %. For PSR1, the prompt mechanism is predominant, followed in importance by the nitrous oxide and NNH mechanisms. In PSR2, the Zeldovich mechanism is most important, followed closely by the nitrous oxide mechanism.

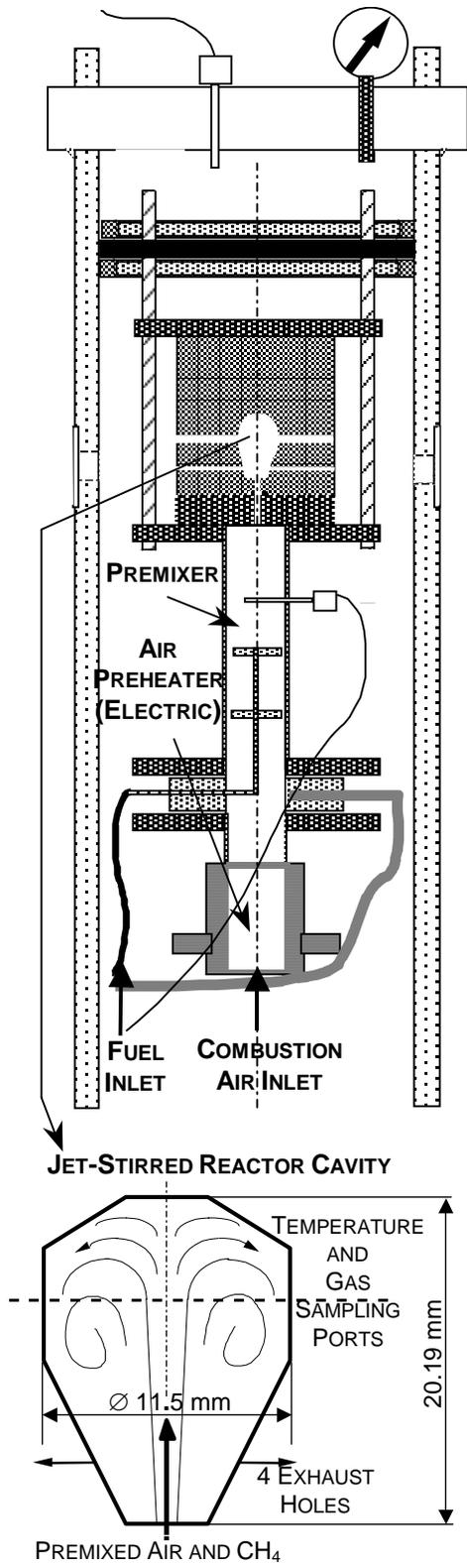
Conclusions

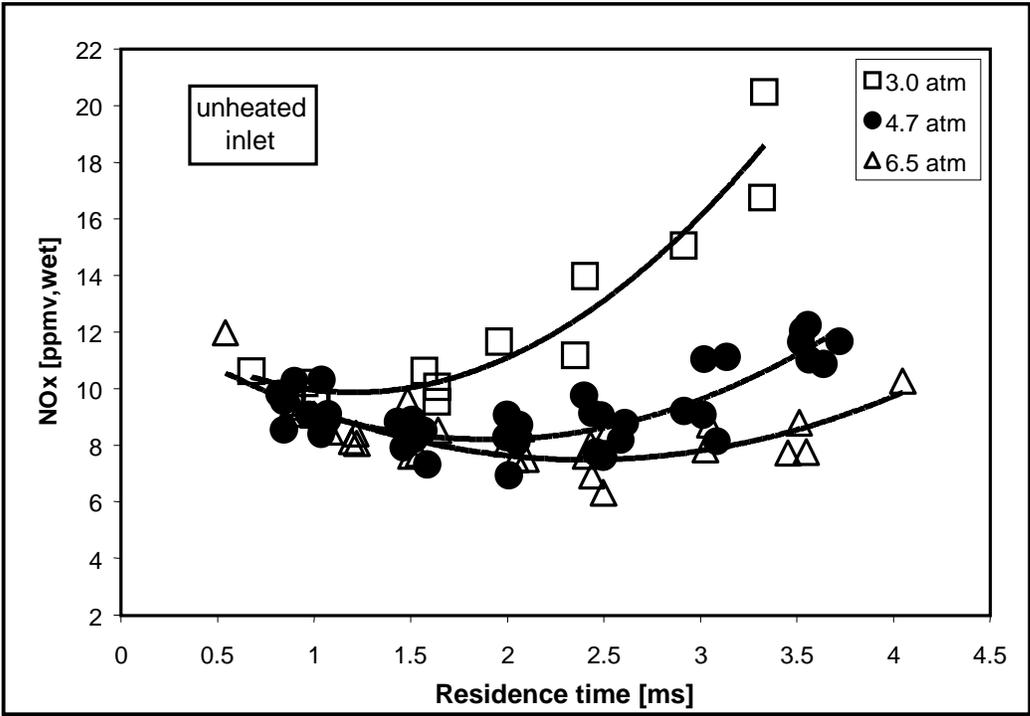
The present research provides new experimental data on the formation of NO_x in the high-pressure jet-stirred reactor, through the operation of the reactor over an 8-fold change in residence time and a 17-fold change in the Damköhler number (Da). Data are provided on NO_x as a function of residence time, pressure, and inlet temperature. Two PSR modeling is applied to the NO_x and CO behavior in the reactor. With $Da < 0.1$, the NO_x and CO are well modeled by treating the reactor as a single PSR. With $Da > 0.2$, a small first PSR followed by a large second PSR is required to jointly model the NO_x and CO. The first PSR represents the non-equilibrium flame zone, while the second PSR represents the stirred post-flame zone. Turbulent flame length modeling provides an independent methodology for determining the flame zone volume required in the two PSR modeling. For $Da \cong 0.06$ (corresponding to lowest residence time), the turbulent flame thickness is about 5.5 mm, a value similar to the reactor radius. The flame fills the reactor. For $Da \cong 1$ (corresponding to largest residence time), the turbulent flame thickness is about 1.5

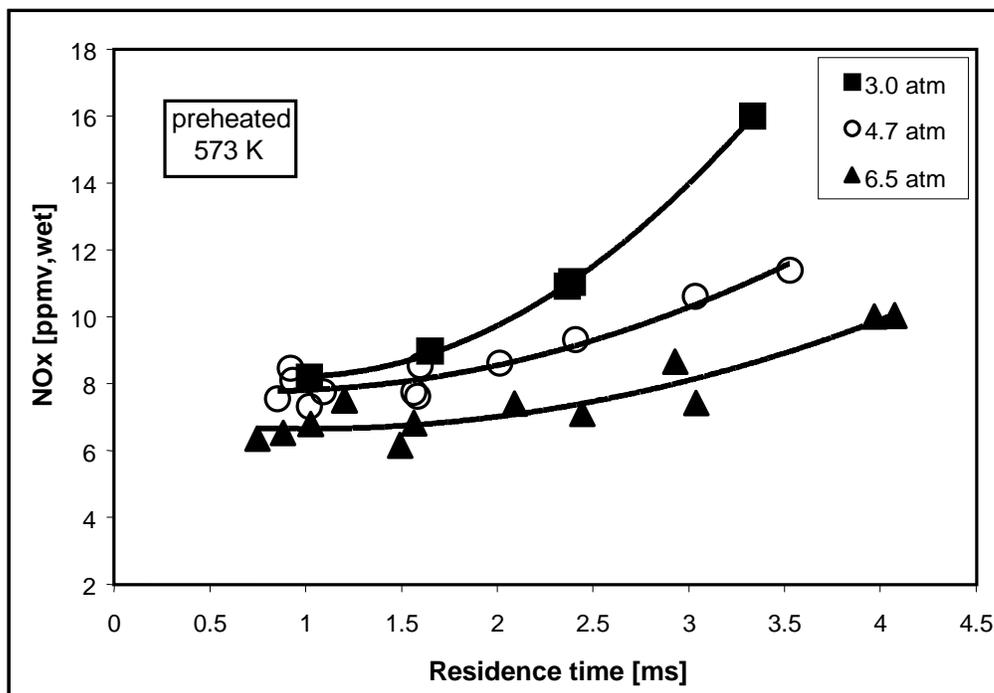
mm, implying a flame volume of few percent of the reactor volume. The significant decrease in the flame volume with increasing Da is consistent with the PSR modeling of the measured NO_x and CO concentrations.

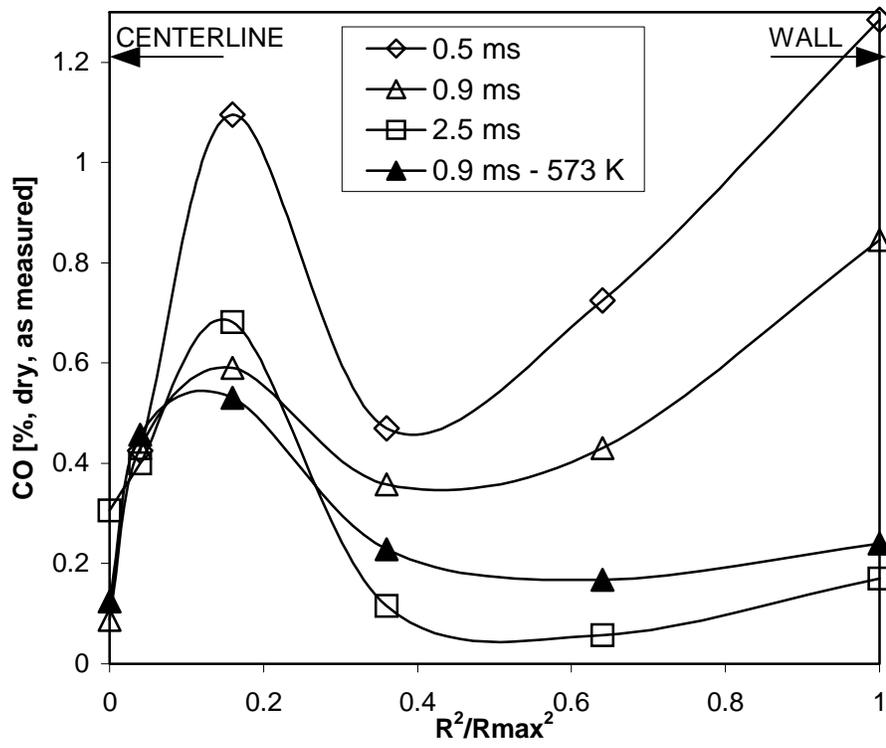
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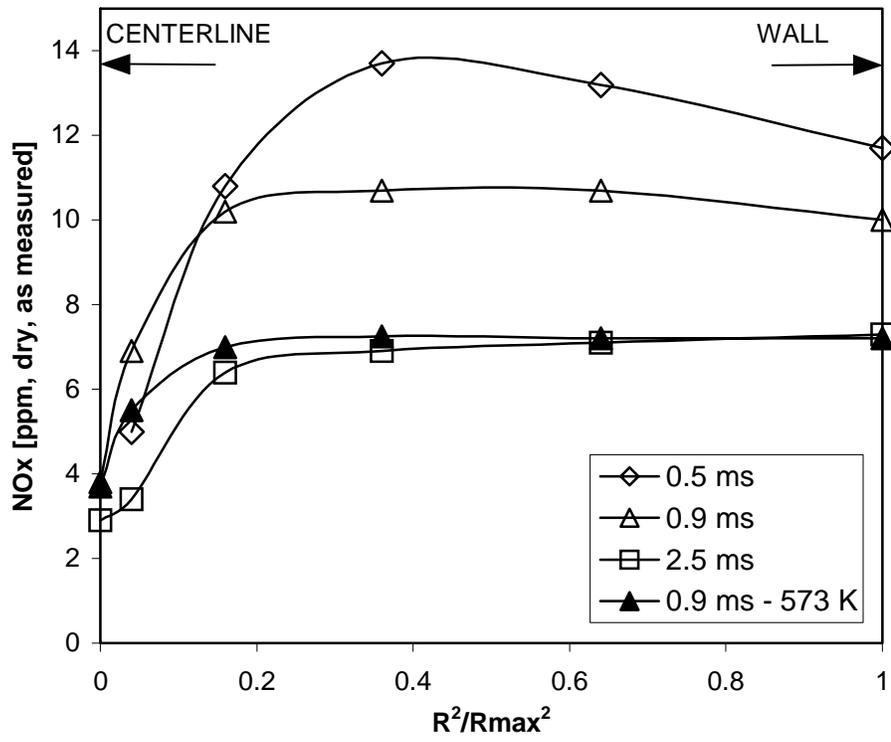
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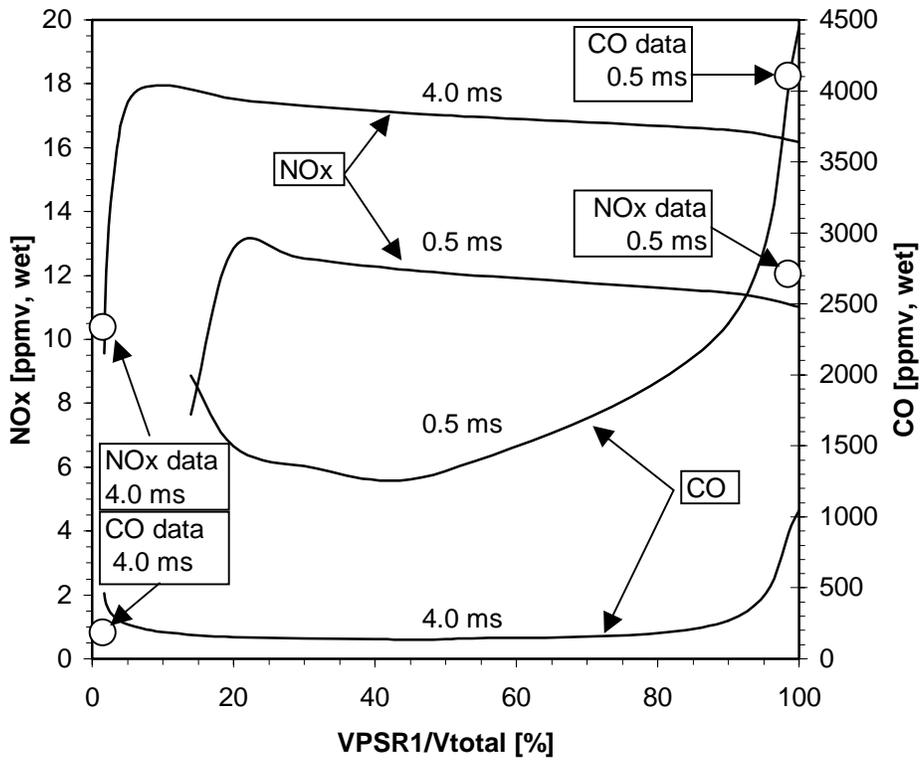












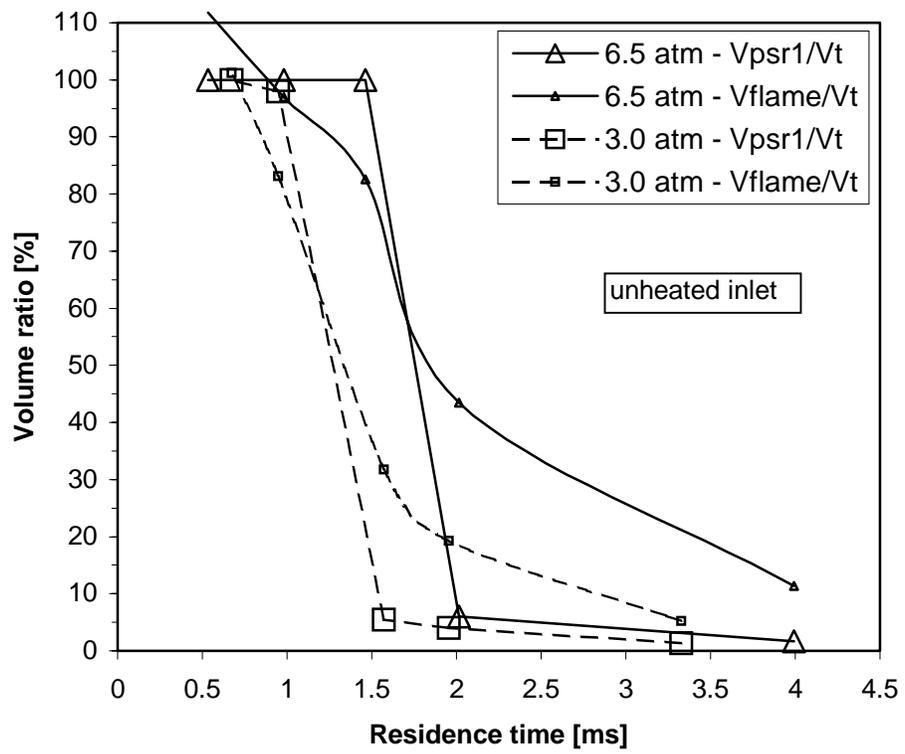


Figure Captions

FIG. 1. High-pressure jet-stirred reactor (HP-JSR) rig (top) with the jet-stirred reactor cavity (bottom). Reactor inlet nozzle diameter is 1.4 mm.

FIG. 2. NO_x versus residence time at 3.0, 4.7 and 6.5 atm for unheated inlet. Mean fuel-air equivalence ratio is 0.71.

FIG. 3. NO_x versus residence time at 3.0 and 6.5 atm for preheated inlet (573 K). Mean fuel-air equivalence ratio is 0.62.

FIG. 4. Spatial profiles of CO across the HP-JSR at $2/3^{\text{rd}}$ reactor height for 6.5 atm and 0.5, 0.9, 2.5 ms residence times with unheated reactants, and for 0.9 ms residence time with reactants preheated to 573 K.

FIG. 5. Spatial profiles of NO_x across the HP-JSR at $2/3^{\text{rd}}$ reactor height for 6.5 atm and 0.5, 0.9, 2.5 ms residence times with unheated reactants, and for 0.9 ms residence time with reactants preheated to 573 K.

FIG. 6. CO and NO_x exiting PSR2 in the two-PSR model, based on GRI-Mech. 3.0. Residence times are 0.5 and 4.0 ms, pressure is 6.5 atm, and there is no preheat. The circles indicate experimentally measured values of CO and NO_x.

FIG. 7. Ratio of PSR1 volume to the total reactor volume (V_{psr1}/V_t) corresponding to the best match between the measured and modeled data, and the ratio of flame volume to the total volume (V_{flame}/V_t) estimated by turbulent premixed flame correlations, plotted versus reactor residence time. The pressures are 3.0 and 6.5 atm and there is no preheat.