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Mechanisms of NO formation in MILD combustion of CH₄/H₂ fuel blends

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ABSTRACT

The mechanisms of formation and destruction of NO in MILD combustion of CH₄/H₂ fuels blends are investigated both experimentally and numerically. Experiments are carried out at a lab-scale furnace with the mass fraction of hydrogen in fuel ranging from 0% to 15%; furnace temperature, extracted heat and exhaust NO_x emissions are measured. Detailed chemical kinetics calculations utilizing computational fluid dynamics (CFD) and well-stirred reactor (WSR) are performed to better analyze and isolate the different mechanisms.

When the MILD combustion of the CH₄/H₂ fuel is established in experiments, the thermal field is quasi uniform and the high temperature zone is located at the junction of the fuel and air jets. As the mass fraction of hydrogen in fuel is increased from 5.7% to 14.4%, although the furnace average temperature is increased, the NO_x emission remains unchanged. This cannot be explained by the thermal NO mechanism. CFD and WSR simulations both suggest that, when equivalence ratio ≤ 0.8, the N₂O-intermediate route controls the NO formation and the NO-reburning reaction is also strong. With the hydrogen addition, the importance of the NNH route is increased but that of the prompt route is decreased, consequently non-affecting the NO_x emission as measured.

Chemical kinetics calculations indicate that the conversion from NO to NO₂ becomes significant and thus the relative importance of NO₂ is increased in the total NO_x emission under low temperature MILD conditions. As the reactor temperature is increased from 1100 K to 1600 K, the importance of N₂O route decreases while that of thermal route increases. In contrast, as the initial mass fraction of oxygen is increased from 3% to 9%, the importance of N₂O route increases but that of the prompt and NNH routes decreases. Likewise, as the equivalence ratio increases, the NO-reburning reaction becomes strong. Worth noting is that the N₂O-intermediate route controls the NO production under fuel lean conditions whereas the prompt route is dominant in rich ones.

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Nomenclature			
Symbols		X_{NO_x}	NO _x emission in volume, ppm
A_i	pre-exponential factor	Y_{CH_4}	initial mass fraction of CH ₄ in the well-stirred reactor, %
C_ξ	the volume fraction constant of eddy dissipation concept model (=2.1377)	Y_{CO_2}	mass fraction of CO ₂ , %
C_τ	the time scale constant of eddy dissipation concept model	$Y_{H_2}^f$	initial mass fraction of H ₂ in fuel, %
D_a	diameter of the air nozzle exit, mm	Y_{H_2}	initial mass fraction of H ₂ in the well-stirred reactor, %
D_f	diameter of the fuel nozzle exit, mm	Y_j	the mass fraction for the jth species, %
K_u	gas recirculation rate	Y_{N_2}	initial mass fraction of N ₂ in the well-stirred reactor, %
p	pressure, atm	Y_{O_2}	initial mass fraction of O ₂ in the well-stirred reactor, %
$Q_{extracted}$	extracted heat, kW	$Y_{O_2}^o$	initial mass fraction of O ₂ in oxidant, %
Q_{input}	thermal input, kW	Greek letters	
T	temperature, K	ξ	characteristic length fraction of fine scales, m
$T_{average}$	average temperature of the furnace centerline, K	τ	residence time, s
$T_{exhaust}$	exhaust temperature, K	ϕ	equivalence ratio
T_{mean}	mean temperature of the entire furnace, K		
T_{WSR}	temperature of the well-stirred reactor, K		

Introduction

Binary fuel blends are widely used in developing high efficiency and low emission combustion devices [1]. Adding hydrogen to methane (or natural gas) can promote ignition and enhance flame stability [2–5], and offer definite advantages in applications such as combustion furnaces and engines [6,7]. However, the hydrogen addition generally increases the flame temperature and thus yields more NO_x emissions. To solve this problem, one of the effective methods is to operate the combustion in moderate or intense low oxygen dilution (MILD) condition to suppress NO_x emissions [8–10].

The MILD combustion is achieved by strong exhaust gases recirculation, where reactants are intensely diluted and hence reactions occur volumetrically without visible flame-front. Relative to the conventional counterpart, the MILD combustion increases the in-furnace thermal uniformity and efficiency, and simultaneously suppresses NO_x emission [11–14]. Although considerable work has been published on MILD combustion [14–28], most of the previous investigations have focused on firing hydrocarbon gaseous fuels or pulverized coal. To understand the characteristics of the MILD combustion of hydrogen containing fuels, both the experimental and numerical investigations are essential.

In Table 1 a summary of published experimental work on the MILD combustion of the CH₄/H₂ fuel in furnaces is presented. Derudi et al. [29,30] showed that, relative to burning pure CH₄, the MILD combustion of the CH₄/H₂ fuel requires higher jet velocity to establish, but it can operate at lower average furnace temperatures. They also found that the addition of hydrogen leads to complete oxidation of the hydrocarbon under MILD conditions. Parente et al. [31] carried out numerical and experimental investigations of the MILD combustion of the CH₄/H₂ fuel (with hydrogen content up to 20% by wt.). For their MILD combustion system, they discovered that the influence of molecular diffusion should be considered in the numerical prediction of H₂ distribution, but its effects on the temperature field and major species are negligible. They also developed a simple NO formation mechanism based on the thermal and prompt routes to predict NO emissions. The MILD combustion in their experiments notably operated in relatively high temperatures (>1800 K). Parente et al. [32] further developed a simplified approach to predict NO formation in the MILD combustion of CH₄/H₂ mixtures. They found that the NNH and N₂O-intermediate routes are crucial when the MILD combustion occurs at low temperatures and hydrogen is added to the fuel. Arghode et al. [33] found that the addition of H₂ results in a slight increase of the NO emission for the same equivalence ratio possibly due to a higher flame temperature. Galletti et al. [34] pointed out

Table 1 – Summary of experimental investigations on the MILD combustion of the CH₄/H₂ fuel in furnaces.

Fuel	Y_{H_2, W_2}^f (%)	T_a (K)	P (kW)	NO _x emissions (ppm)	CO emissions (ppm)	Reference
CH ₄ /H ₂ mixture	5, 15	≥1173	0.2–0.3	<30	<50	[29]
CH ₄ /H ₂ mixture	15	≥1173	0.2–0.3	<30	<50	[30]
CH ₄ /H ₂ mixture	5.5	800–900	13	59–105	N/A	[31]
CH ₄ /H ₂ mixture	50	N/A	10	25–45	N/A	[32]
CH ₄ /H ₂ mixture	8	300	6.25	0–30	20–600	[33]
CH ₄ /H ₂ mixture	15	1223, 1312	0.2–0.3	17	<50	[34]
CH ₄ /H ₂ mixture	0–100	298, 858	20	8–16, 26–42	3–60, ≤10	[35]

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