



MILD oxy-combustion of gaseous fuels in a laboratory-scale furnace

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ABSTRACT

The present study investigates the characteristics of Moderate or Intense Low-oxygen Dilution (MILD) oxy-combustion in a laboratory-scale furnace. Experiments using natural gas (NG), liquefied petroleum gas (LPG) and ethylene (C₂H₄) are carried out at a firing rate of 13 kW. The furnace temperatures and exhaust emissions are measured for a range of equivalence ratios and external-CO₂ dilution rates.

It is observed that MILD combustions occur for the three fuels even when using pure oxygen as oxidant. When diluting oxidant by CO₂ at a fixed rate, the MILD combustion can be established as long as the equivalence ratio (Φ) is sufficiently high. The region of MILD combustion is found to be wider with dilution by CO₂ than by N₂. Notably, also, the operating range of MILD combustion is larger for NG than LPG or C₂H₄ as fuel.

Moreover, when $\Phi < 1$, as Φ is increased, the furnace temperature rises slightly but the NO_x emission decreases. This cannot be explained when using the traditional thermal NO_x mechanism. Indeed, using various NO mechanism models, our calculations show very low NO emissions resulting from the thermal, prompt and NNH routes but a much higher value from the N₂O-intermediate route. Namely, only the latter mechanism plays a crucial role in forming NO. Also important is that the NO reburning appears to reduce NO emissions notably and so should not be ignored in the MILD combustion.

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1. Introduction

Oxy-combustion has generated significant interest for carbon capture and storage (CCS) [1–3]. This technology also has additional benefits such as emission reduction and lower costs of flue gas cleanup [4–6]. By eliminating nitrogen from the combustion medium, the flue gas will consist mainly of CO₂ and H₂O. Recirculated combustion products are used for diluting a nearly pure O₂ stream. Due to the higher heat capacity and radiative properties of CO₂ compared to N₂, an increased initial oxygen concentration (23–35%) for this combustion is required to achieve gas temperatures and heat transfer performances similar to those in air–fuel combustion [7,8]. The flue gas can then undergo a condensation process to remove H₂O to end up with a flue gas that consists almost entirely of CO₂. The nearly pure CO₂ product stream obtained via this process is suitable for use in enhanced oil recovery (EOR), coal bed methane (CBM) production, or geologic sequestration [9]. More recently, this technology has been adopted to substitute the original integrated gasification combined cycles (IGCC) plan in the US DOE FutureGen 2.0 program [10]. Details on the characteristics [4,6,11,12] and combustion process [4], as well as recent developments in pilot-scale and commercial-scale

demonstration plants [13] of the oxy-fuel combustion, can be found in the above mentioned articles.

While successful, the technology still faces many challenges [4]. At least three features of the oxy-fuel combustion process need to be better understood: (a) the approach of further increasing the thermal performance of the system, (b) the subsequent potential of NO_x forming, due to leakage of air (air-ingress) or nitrogen in the fuel itself [5], and (c) the challenge of improving the stabilization of oxy-fuel combustion due to the lower adiabatic flame temperature, delayed ignition and lower burning rate in a CO₂ diluted environment [4,14–16]. However, the increase of the flame temperature and combustion intensity, which is one of the approaches for further increasing the thermal performance, often comes at the price of high pollutant emissions (e.g., NO_x) [17]. Although NO_x emissions can be controlled by air or fuel staging, or low-NO_x burner technology, the flame temperature is also decreased undesirably. It is difficult to simultaneously satisfy the requirements of high efficiency and low pollution in oxy-fuel combustion process. One way of addressing this is to use the principle of MILD combustion.

MILD combustion is an efficient and clean combustion technology [18–26]. Extremely low NO_x emission and simultaneously high thermal efficiency can be achieved in the MILD combustion process. To our best knowledge, the three groups, i.e., Wüning and Wüning [20], Nippon Furnace Kogyo Kaisha Ltd. (NFK-Japan)

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Nomenclature

Symbols

A_o	area of the nozzle exit (m^2)
c_p	volumetric heat capacity at constant pressure ($kJ/(m^3 K)$)
D_{ex}	diameter of the exhaust outlet (mm)
D_f	diameter of the fuel nozzle exit (mm)
D_o	diameter of the oxidant nozzle exit (mm)
h	lower heating value of the fuel (kJ/kg)
J	jet momentum rate ($kg m/s^2$)
J_f	fuel jet momentum rate ($kg m/s^2$)
J_o	oxidant jet momentum rate ($kg m/s^2$)
K_v	relative recirculation rate
K_v^*	external relative dilution rate
M_d	total mass flow rate of diluents (kg/s)
M_e	mass flux of the external diluents (kg/s)
M_f	fuel jet mass flux (kg/s)
M_i	mass flux of internal entrained exhaust gas (kg/s)
M_o	oxidant jet mass flux (kg/s)
m_o	jet mass flux (kg/s)

P	thermal input power rate (kW)
T	furnace temperature (K)
T_p	preheat temperature of oxidant
T_{ad}	adiabatic flame temperature (K)
T_{max}	maximum temperature (K)
T_w	furnace wall temperature (K)
U_o	velocity of the initial reactant
$Y_{CO_2}^f$	initial mass fraction of CO_2 in the fuel jet (%)
$Y_{CO_2}^o$	initial mass fraction of CO_2 in the oxidant jet (%)
$Y_{O_2}^o$	initial mass fraction of O_2 in the oxidant jet (%)

Greek letters

λ	thermal conductivity ($W/(m K)$)
α	mass diffusivity in air (m^2/s)
ν	kinematic viscosity (m^2/s)
ρ_o	density of the initial reactant (kg/m^3)
Φ	equivalence ratio

[19,21] and the International Flame Research Foundation (IFRF) [22,23], have originally been involved in the development of this technology. In the German path [20,24], from the early 1990s, this technology was named as flameless oxidation (FLOX). In the period from 1992 to 1999, the NFK-Japan and the IFRF worked together and the Japan-IFRF design [19] was distinct from the design of Wüning and Wüning [20]. The IFRF reports, e.g., [25], in the period from 1992 to 1999 and the journal publications, e.g., Weber et al. [23], described the experimental and numerical results. Later, Cavaliere and Joannon et al. [18,26] introduced the acronym MILD for the technology, which has been well recognized by the international combustion community.

The main advantages of MILD combustion are uniform temperature distribution, increased net radiation flux, extremely low NO_x emissions and the stable operating of the combustion mode without flame stabilization problem [20,27–33]. Theoretically, if the oxy-fuel technology is combined with the MILD technology to achieve the MILD oxy-fuel combustion, the thermal efficiency may be improved and simultaneously the NO_x emission may be suppressed. Hence, the MILD oxy-combustion may have the potential to offer more advantages over the standard oxy-combustion [4].

Although a good volume of work has been carried out in the separate field of the MILD combustion [28–33] and the oxy-fuel combustion, the information on the combination of the MILD combustion and oxy-fuel combustion is limited. The IFRF tested MILD oxy-NG combustion in a project called OXY-FLAM [34,35] in the period from 1995 to 1999, although without CO_2 recirculation. The project dealt both with the detailed characterization of oxy-combustion burners and with the validation of new computer codes as sub-models for oxy-NG combustion calculations. The experimental work included input–output and measurements in flames in the thermal input range 0.7–1.0 MW. They found good predictions by using the eddy dissipation concept mode with the full equilibrium chemistry procedure [34,35]. Blasiak et al. [36] applied the oxy-NG combustion to thermal treatment processes of wastes and the recovery of zinc bearing feed on a rotary kiln. They found that the MILD oxy-combustion not only increases the productivity of the rotary kiln but also reduces the fuel consumption and NO_x emissions. Krishnamurthy et al. [37] compared conventional and MILD oxy-combustion under similar conditions using propane as fuel (200 kW). They found that the MILD

oxy-combustion can be achieved by asymmetric injection of high velocity oxygen (at near sonic velocities). They also found that the soot formation was negligible in MILD and was higher in conventional flame mode. Krishnamurthy et al. [38] also found that the NO_x emission of MILD oxy-combustion can be maintained at very low levels and is insensitive to air-ingress. The air leakage was indicated by the amount of oxygen in the flue gases. For conventional flame combustion, the NO_x emission increases as oxygen level of the exhaust increases. Their test was performed in a pilot-scale furnace ($8 m^3$) where air-ingress into the combustion chamber was simulated by leaking air into it, in order to raise the free oxygen content in the combustion gases. The oxygen content was measured in the furnace flue gas outlet. Stadler et al. [39] developed a high speed injection MILD oxy-combustion 100 kW burner. Their experimental setup has successfully achieved stable MILD oxy-combustion with 15% oxygen volume concentration as compared to 17% with a standard swirl burner. Stadler et al. [40] also found that the flue gas recirculation led to a reduction of NO_x emissions of up to 50% for the swirl flame, whereas in MILD combustion this reduction is around 40% compared to CO_2/O_2 . Heil et al. [41] presented an experimental investigation of methane MILD oxy-combustion with different inert gases (N_2/O_2 and CO_2/O_2) and O_2 concentrations (21 vol% and 18 vol%). The result demonstrated that by eliminating the influences of molar heat capacity, CO_2 dissociation and thermal radiation, the effects of high CO_2 concentrations on combustion rates can be attributed to its participation in the chemical reactions. An increase of O_2 led to a reduction of this impact. Despite the above investigations, the basic information on the MILD oxy-combustion is still sparse and calls for more research.

To help address the above deficit, the present study is designated to investigate the detailed performance and stability characteristics of a parallel jet burner system operating at a laboratory-scale MILD combustion furnace burning NG, LPG and C_2H_4 with air or O_2/CO_2 mixture. This is achieved through the measurements of in-furnace temperatures, exhaust emissions and stability limits. The NO_x formation mechanisms are investigated from the computational fluid dynamic (CFD) simulation. Effects of the initial mass fraction of CO_2 , equivalence ratio, burner configuration, fuel type, and thermal field are also discussed. Finally, the stability limits of MILD oxy-combustion at different oxidant mixtures are presented with discussion.

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