



Carbon dioxide diluted methane/oxygen combustion in a rapidly mixed tubular flame burner



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ABSTRACT

An inherently safe technique of rapidly mixed tubular flame combustion, in which fuel and oxidizer are separately injected from their own slits, has been extended to CO₂ diluted methane/oxygen combustion. Two different cases are investigated. When CO₂ is added only to the oxidizer slit, a stable tubular flame is obtained for a wide range of equivalence ratios with the oxidizers of oxygen mole fraction between 0.21 and 0.50. However, once the oxygen mole fraction exceeds 0.60, the stable flame is obtained within a small equivalence ratio range near the lean extinction limit, and the stable range becomes narrower and narrower with increasing the oxygen mole fraction. When CO₂ is added also to the fuel slit so as to maintain the oxidizer/fuel injection velocity ratio near unity, and the flow rates of fuel and oxidizer are increased, the stable range is dramatically widened and a stable flame at stoichiometry can be obtained up to the oxygen mole fraction of 0.86, which yields adiabatic flame temperature around 2950 K. The Damköhler number, defined as the mixing to reaction time ratio, is examined to discuss the criterion for the establishment of stable tubular flame. It is found that when CO₂ is added only to the oxidizer slit, stable flame can be obtained for $D_a < 1$, whereas not for $D_a > 1$ due to formation of diffusion flames near the fuel slits. When CO₂ is added also to the fuel slit, however, stable flame can be established even for $D_a \geq 1$ as well as $D_a < 1$. Detailed observations indicate that when D_a is adequately smaller than unity, the flame is uniform in luminosity, whereas the flame is non-uniform when D_a is near or larger than unity. It is considered that the diffusion flame formation at the fuel slits is prohibited due to the high injection velocities, which gives a chance of fuel/oxidizer mixing, however, the tubular flame obtained is of non-uniform structure due to insufficient mixing time when $D_a \geq 1$.

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1. Introduction

In the view of increasing concerns on the environmental issue and energy security all over the world, oxy-fuel combustion, in which combustion takes place in a CO₂ environment, has attracted considerable attention as a promising approach for low-cost CO₂ capture and sequestration (CCS) combined with reduction of NO_x emissions [1,2].

During the past decades extensive studies have been conducted on oxy-fuel combustion, however, they are concentrated mostly on coal combustion [2–4]. Oxy-fuel combustion of coal has been proposed for a boiler cycle in power generation [3,4], and also fundamental aspects such as flame propagation velocity and lean inflammability limits are investigated for oxy-fuel combustion of coal [5].

Natural gas also receives attention with regard to low CO₂ emission relative to the heat of combustion. Liu et al. [6] numerically investigated oxy-fuel combustion of methane with detailed thermodynamics and chemical kinetics, and provided optimal conditions of oxygen/carbon dioxide ratio in a gas turbine model combustor up to the oxygen mole fraction of 0.5. Amato et al. [7] numerically investigated oxy-fuel combustion of methane, and in addition, experimentally measured CO and O₂ emissions with using a swirl stabilized combustor, to discuss the dependence of the CO and O₂ emissions on stoichiometry, pressure, CO₂ dilution and residence time. Using also a swirl type burner (DLR dual swirl burner), Kutne et al. [8] experimentally investigated oxy-fuel combustion of methane and reported that stable combustion can be obtained in a limited range of oxygen mole fraction between 0.23 and 0.4.

In most oxy-fuel combustion of methane, the oxygen mole fraction is limited less than 0.5. This is partly because high oxygen concentrations are not necessary or inhibited to protect furnace material in power plant systems. However, it had better to develop

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a burner system, which can treat higher oxygen concentration, hence high temperature, which is in great demand in industries such as glass and cement manufacturing.

Although using the similar swirling flow, a distinct different mode of combustion has been developed for methane/oxygen combustion by Shi et al. [9,10]. They use a so-called rapidly mixed tubular flame burner, in which fuel and oxidizer are separately injected into the combustion tube through their own slits. Flame flash-back will never occur [11]. In addition, this burner gives a well-controlled combustion because the flow is laminarized due to strong swirl and large density change with combustion. Furthermore, due to the thermal as well as aerodynamic advantages [12,13], a very wide stable flame region can be obtained.

Although the stable tubular flame range in equivalence ratio becomes narrower once the oxygen mole fraction exceeds 0.5 in the O_2-N_2 oxidizer, a recent reexamination on the methane/oxygen combustion suggests that stable flame range can be expanded by setting the oxidizer/fuel injection velocity ratio near unity [10].

In this study, the inherently safe technique of rapidly mixed tubular flame combustion has been extended to CO_2 diluted methane/oxygen combustion. To widen the stable flame range, CO_2 is added also to the fuel slit in addition to the oxidizer slit to approach the same injection velocities from the fuel and oxidizer slits. The flame characteristics such as stable range and flame structures have been determined under various oxygen mole fractions and flow rates. Numerical calculation with the Chemkin code is also conducted to discuss the flame characteristics obtained and the criterion for the establishment of stable tubular combustion.

2. Experimental and numerical methods

Figure 1 shows the schematic drawings of the rapidly mixed tubular flame burner. The burner is made of stainless steel, which consists of four tangential slits (two horizontal oxidizer slits and two vertical fuel slits) and a downstream quartz tube. A quartz window is installed at the closed end to permit viewing and photographing the flame front. The inner diameter of the burner is 16 mm, and the slit width (W) and length are 1 and 8 mm

respectively, giving a swirl number of 12.56 [13]. Three combustion tubes with lengths of 100, 75 and 50 mm are used.

The fuel used is methane while the oxidizer is O_2-CO_2 mixture or pure O_2 . For comparison, combustion tests with the oxidizer of O_2-N_2 mixture (oxygen-enriched air) are also conducted. The fuel is injected through the fuel slits while the oxidizer of O_2-CO_2 mixture is individually injected from the oxidizer slits, as shown in the left illustration of Fig. 1(b), which is denoted as Case I. Two specific examples are given in Table 1 (cases ① and ②), in which the flow rates of O_2 and CO_2 in the oxidizer slit are denoted as Q_{O-O_2} and Q_{O-CO_2} , respectively, while the flow rate of CH_4 in the fuel slit is denoted as Q_{F-CH_4} . Here Q is the standard-state volumetric flow rate with unit of m^3/h . The total flow rate in the oxidizer slit is denoted as $Q_{O-total} = Q_{O-O_2} + Q_{O-CO_2}$, while the total flow rate in the fuel slit is denoted as $Q_{F-total}$, which is equal to the fuel flow rate Q_{F-CH_4} in Case I. The oxygen mole fraction β is defined as $\beta = Q_{O-O_2} / (Q_{O-O_2} + Q_{O-CO_2})$. In the case of O_2-N_2 mixture, Q_{O-CO_2} is replaced by Q_{O-N_2} . The equivalence ratio ϕ is the overall value for the rapidly mixed combustion assuming complete mixing.

In order to increase the fuel injection velocity, the CO_2 diluent is added also to the fuel slit, as shown in the right illustration of Fig. 1(b), which is denoted as Case II. Corresponding to Case II, several specific combustion tests are also given in Table 1 (cases ③–⑨). The flow rates for the CO_2 diluents in the oxidizer and fuel slits are denoted as Q_{O-CO_2} and Q_{F-CO_2} , respectively. The total flow rate in the oxidizer slit is $Q_{O-total} = Q_{O-O_2} + Q_{O-CO_2}$, and that of the fuel slit is $Q_{F-total} = Q_{F-CH_4} + Q_{F-CO_2}$ in Case II. Here we assume a mixture of O_2 and CO_2 from the oxidizer slit and CO_2 from the fuel slit, and define the oxygen mole fraction β' in the assumed mixture, denoted as $\beta' = Q_{O-O_2} / (Q_{O-O_2} + Q_{O-CO_2} + Q_{F-CO_2})$. To further increase β' at the stoichiometric condition, in cases ⑥–⑨ in Table 1, all the CO_2 diluent is added to the fuel slit.

The flame is photographed using two conventional digital cameras to obtain the front view through the quartz window and the side view through the quartz combustion tube simultaneously. To investigate the flame structure in more detail, chemiluminescence of CH^* and OH^* radicals is determined using a high speed video camera FASTCAM-APXRS (Photron Co.) with HS-IRO (LaVision Inc.) intensifier and filters. The center wavelength and half-width of the CH^* filter are 433.0 and 1.6 nm, and those of the OH^* filter are 313.0 and 10.0 nm, respectively. An optimum gain value of 75 μs , a frame rate of 10,000 frames/s, a resolution of 512×512 pixels, and an aperture of 4.5 are utilized to detect the CH^* and OH^* chemiluminescence.

To quantitatively analyze the criterion for tubular flame establishment, the Damköhler number, defined as the mixing to reaction time ratio, has been examined. The chemical reaction time of CH_4/O_2-CO_2 mixture is calculated by using the Chemkin Premix code [14,15] in the Chemkin-PRO software, in which the GRI-Mech version 3.0 [16] is used as the reaction mechanism. Initial temperature of 298 K, pressure of 1 atm and composition of the unburned mixture are assigned to define the inlet boundary condition. It is specified that all gradients vanish at the exit boundary. The mixing time is estimated based on the previous study [10].

3. Results

3.1. Case I: CH_4/O_2-CO_2

At first, observations were made for Case I, and combustion tests under various oxygen mole fractions were attempted.

For conventional oxy-fuel combustion of methane, the operating oxygen mole fraction is usually above 0.21 [8,17–19], since it is difficult to establish the stable combustion owing to very low burning velocity [20] and also low flame temperature. In this study

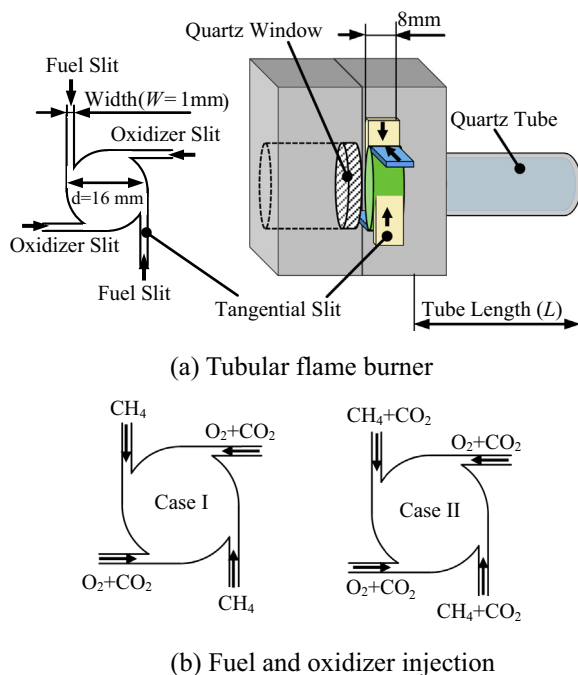


Fig. 1. Schematics of the tubular flame burner.

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