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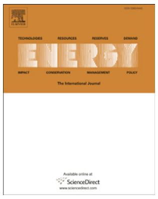

Comparative study on the effects of nitrogen and carbon dioxide on methane/air flames

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ABSTRACT

The measurement of flame temperature profile and flame computation were conducted to analyze the effects of carbon dioxide and nitrogen dilution on methane/air flames. Results indicated that carbon dioxide addition shows more significant effects on the thermal properties of flame, except for flame thickness. Flame kinetic analyses were implemented for the heat release of individual reaction. It is suggested that the heat release contribution of dominant reaction is primarily affected by flame stoichiometry, while the dilution shows weak effect. The concentrations of reactive species play important influences on the progress rate of reaction. The peak product of reactive species concentrations of the dominant reactions could be approximated by an exponent expression of the concentration of initial reactants and dilution rate, and a greater exponential index was obtained for the flames with carbon dioxide addition. The emission indices and combustion efficiency were evaluated to assess the profit in NO suppression and the loss in combustion deterioration. Results indicated that the profit-loss ratio is influenced by both flame equivalence ratio and the component of diluent gas. For rich methane/air flame, nitrogen dilution shows a greater value, while a larger benefit is obtained for stoichiometric flame diluted by carbon dioxide.

1. Introduction

Nitrogen oxides (NOx) plays an important role in forming particulate matter, ground level ozone and acid rain, which are harmful to both human health and environment, therefore the reduction of NOx emission, particularly from industrial sources such as the combustion of hydrocarbon fuel, has received much attention. Up to now, some new combustion techniques, e.g. flameless combustion in industrial furnaces and EGR (exhaust gas recirculation) in engines, have been widely applied to suppress NOx production. Flameless combustion [1] also known as HiTAC (high temperature air combustion), MILD (moderate or intense low oxygen dilution) combustion or CDC (colorless distributed combustion), and the combustion of exhaust gas recirculation [2]are both based on internal flue gas recirculation. Their intrinsic nature is dilution combustion, in which, the reintroduction of hot exhaust gas into fresh fuel/air mixture would reduce the oxygen concentration in the flame zone to decrease the reaction rate of flame and absorb part of reaction heat of the flame, due to the chemical inertness of diluent gas. As the result, the peak temperature of flame would be decreased or the temperature peak would be eliminated through having a nearly homogeneous combustion in combustion chamber, and ultimately, ultra-low NOx combustion is achieved.

Owing to the remarkable advantage of dilution combustion in inhibiting NOx formation, the effects of diluent gas on the combustion characteristics of hydrocarbon fuels have been extensively studied. Plessing, Peters and Wünning [3] made an experimental investigation on the flameless combustion in a combustion chamber with highly preheated air and strong exhaust gas recirculation. They found that the dilution of hot exhaust gas can result in the low flame temperatures even under significant air preheating condition. Maiboom, Tauzia and Hétet [4] examined the effects of cooled EGR on combustion and NOx/PM emissions in an automotive diesel engine under low-load and part load conditions. Their results

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reported that the properties of the diluent gas in mixture have significant influences on engine characteristics.

As we know, the burned exhaust gas of hydrocarbons combustion in air primarily consists of nitrogen, carbon dioxide, as well as water vapor. In order to distinguish and quantify the component effects of diluent gas, numerous numerical and experimental studies were implemented for hydrocarbon flames with monocomponent diluent gas. Coppens, Deruyck and Konnov [5] studied the effects of nitrogen on the NO formation in methane/hydrogen flames and reported that nitrogen dilution could decrease the concentration of NO in the flames at any equivalence ratios. Miao et al. [6] and Vancoillie et al. [7] measured the laminar flames of methane and methanol in air with nitrogen dilution and reported that the flame burning velocities decreased with the increase of nitrogen concentration. Prathap et al. [8] studied the effects of nitrogen on the laminar combustion of syngas fuel. Lee et al. [9] studied the effects of carbon dioxide dilution on NOx emissions and combustion instabilities in lean premixed flames and pointed out that the flame temperature declined with the increase of the dilution rate of carbon dioxide. Samanta and his co-workers [10] studied the effect of carbon dioxide on the flame structure of methane/air mixture. Hinton and Stone [11] measured the laminar burning velocities of methane flames in air with carbon dioxide addition at the elevated temperatures and pressures, and the disadvantage of carbon dioxide on reducing flame oxidation rate was quantified.

Heil et al. [12] experimentally studied on the dilutions of nitrogen and carbon dioxide on the stable methane flameless combustion by in-furnace measurements of combustion product compositions and temperatures. Their results suggested that the chemical effects of carbon dioxide are more significant than that of nitrogen on the production and consumption rates of CO in oxy-fuel combustion. Zhang et al. [13] made an investigation on the different effects of nitrogen and carbon dioxide by experimental observation in a furnace of 20 kW and chemical kinetics calculation of a well-stirred reactor. They found that the requirement for establishing MILD combustion is less stringent with dilution by carbon dioxide than by nitrogen. Several studies [14–17] have suggested that both nitrogen and carbon dioxide can significantly inhibit NO production and show the different impact on the fuel conversion efficiency, due to the differences in their thermophysical, radiative and chemical properties. As water vapor is also one of the most important components in the actual diluent gas, the influence of water vapor addition on hydrocarbon combustion has also been compared with that of carbon dioxide previously [18,19].

Exploring the microscopic chemical processes of combustion is essentially useful to find the actual effects of diluent on hydrocarbon oxidation. The measurements and computations of flame structure have been considered a shared method for flame chemical processes research. Flame structure is a comprehensive term. One of the most widespread descriptions of flame structure is to measure or compute the reactive species profiles of flame. The analyses of flame species distribution have been invariably implemented for finding the key radicals and dominant reactions that underlie and sometimes control the chemical reaction routes of hydrocarbon oxidation [20,21]. Another important form of flame structure is the thermal structure, which is always related to the determination of flame heat release rate. Lafay et al. [22] carried out thermal structure analyses for methane/air enriched by hydrogen. The reaction contribution was computed to quantitatively interpret the decrease of the flame thickness of hydrogen enriched flames. Hu et al. [23] and Li et al. [24] computed the heat release rates of premixed methane/hydrogen/air flames, and the key radicals and reactions that underlie and control the chemical reaction routes were found.

The flames of most of gaseous and liquid fuels diluted by nitrogen and carbon dioxide had been extensively studied and the effects of nitrogen and carbon dioxide dilution on the combustion were well-explored. However, most of previous studies focused on applications in developing low NOx combustion techniques or determining the fundamental properties, such as flame burning velocity and mixture flammability etc. Very few works are relevant to the study on the thermal structures of flame. In order to gain insight on the kinetics changes occurred in reaction routes of diluted flames, further investigations are still necessary.

This work aims to provide a comparative analysis on the combustion properties of premixed methane/air flames diluted by nitrogen and carbon dioxide. The flame temperature profile was determined by the traveling thermocouple approach applied on McKenna flat flames. Flame computation was implemented by solving the species conservation equations of a burner-stabilized flame on the basis of the measured flame temperatures. The global thermal properties of flame were computed in terms of flame temperature, flame thickness and heat release rate. Reaction contribution in heat release, emission indices and combustion efficiency were evaluated to assess the different impacts of nitrogen and carbon dioxide on methane/air flame as well.

2. Experimental setup

In the experiments, one-dimensional premixed laminar methane/air/diluent flames were stabilized on a McKenna flat burner. The experimental setup consisted of two systems for regulating fuel/air supply and measuring flame temperature profile, and a McKenna flat flame burner. Fig. 1 shows a schematic diagram of the experimental setup. The function of the fuel supply system is to regulate the mass flow rates of fuel, air and diluent gas, and provide conditions for gas mixing. The tested fuel was methane with purity above 99.99%. The diluent gas was nitrogen or carbon dioxide with same purity of 99.9%. Methane, diluent gas, and compressed air were introduced into a mixing bomb according to given proportions. Their respective mass flow rates were measured and regulated by mass flow controllers (SIARGO, Model MF5706) with a reported accuracy of 2% of full scale. These gases were mixed in the mixing chamber. After uniformly mixing, the mixture passed into a pressure gauge for stabilizing mixture pressure, and then into the McKenna flat burner to burn. The traveling thermocouple approach [25,26] was well used to measure flame temperature profile. Herein, a fine Pt/13%-Rh thermocouple with the wire diameter of 0.025 mm was fixed in a ceramics cannula with the diameter of 0.8 mm and suspended above the center of the burner. In the measurements, the thermocouple was vertically driven by a motor to pass through the flame zone with a speed of about 0.5 mm/min. The measured temperatures were then corrected according to a standard temperature correction method in which radiation losses from the thermocouple were considered [27]. The surface of the McKenna flat flame burner (the diameter of 60 mm) was made up of a sintered bronze plate with numerous fine meshes to ensure a laminar flow at the outlet of burner. A cooling circuit was nested in the burner body to maintain the constant temperature of the burner plate. In the experiments, an annular nitrogen jet was used as an inert shielding gas to encircle the measured flame for avoiding the diffusive behavior of combustible mixture with ambient air, and the body of the burner was also surrounded by a Pyrex glass to avoid the aerodynamic effect on flame stabilization. In Fig. 1, the dashed line shows the flow direction of the inert shielding gas, and the dotted line is for that of cooling water.

The measurements were conducted at the atmospheric conditions of 300 K and 0.1 MPa, for mixtures with a constant bulk flow rate in volume. In the experiments, 27 methane/air flames were