



# Catalytic combustion of premixed methane/air in a two-zone perovskite-based alumina pileup-pellets burner with different pellet diameters



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## HIGHLIGHTS

- Flame stability limits expanded by depositing perovskite on inert alumina pileup-pellets.
- Flame, flue gas temperatures reduced by catalytic combustion or increasing pellet diameter.
- Unburned hydrocarbon emissions of catalytic were less than 550 ppm and lower than inert.
- CO emissions of catalytic reduced (<20 cm/s) and were kept zero emission levels (>20 cm/s).
- NO<sub>x</sub> emissions of catalytic were less than 2 ppm, whereas inert ranging from 4 to 13 ppm.

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## ABSTRACT

In this study, methane/air combustion in a two-zone catalytic alumina pileup-pellets burner with equivalence ratios varying from 0.55 to 0.70 was researched. The catalytic alumina pileup-pellets were designed by directly depositing perovskite LaMn<sub>0.4</sub>Co<sub>0.6</sub>O<sub>3</sub> on inert alumina pileup-pellets supports. Flame stability limits, pressure drops, flame temperatures, flue gas temperatures, and pollutants emissions of both catalytic and inert burners with different pellet diameters were investigated. The flame stability limits expanded as equivalence ratio or pellet diameter increased. The flame stability limits were also increased by depositing the perovskite catalyst. The pressure drops of catalytic burners were lower than those inert burners in both reaction and cold flows. More uniform axial temperature distributions were obtained for catalytic burners than for inert burners. Flame temperatures and flue gas temperatures were significantly reduced for catalytic burners compared with inert burners, as were pollutants emissions for unburned hydrocarbon (HC), carbon monoxide (CO), and nitrogen oxides (NO<sub>x</sub>). The HC emissions reduced as either pellet diameter or inlet velocity increased. The HC emissions of catalytic burners (less than 550 ppm) were significantly lower than those inert burners (ranging from 900 to 2750 ppm). The CO emissions of catalytic burners reduced significantly at lower velocities ( $S < 35$  cm/s) and were almost unchanged at a level below 50 ppm, at higher velocities ( $S > 35$  cm/s). The CO emissions of catalytic burners were sensitive to changes in pellet diameters at lower velocities ( $S < 20$  cm/s). The NO<sub>x</sub> emissions of catalytic burners (less than 2 ppm) were significantly lower than those inert burners (ranging from 4 to 13 ppm).

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## 1. Introduction

Methane is a major component of natural gas, which is an important resource used worldwide to provide energy. Complete combustion of methane operates at high flame temperatures and leads to the formation of considerable amounts of nitrogen oxide (NO<sub>x</sub>), carbon monoxide (CO), and unburned hydrocarbon (HC).

Therefore, the rapidly increasing need for energy has sparked an interest for methane combustion at lower flame temperatures with higher combustion efficiencies, lower emissions, and flameless combustion modes [1].

Inert porous media combustion can generally fulfill these requirements. Inert porous medium burners operate on the principle that the premixed fuel/air mixture burns within the cavities of a solid porous matrix with superior heat transfer properties for internal heat recirculation. Various studies on the combustion characteristics of porous media burners have been conducted on

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evaluating the effects of burner configurations, excess operating air, and equivalence ratios on combustion performance. Yu et al. [2] reported the combustion characteristics of metal fiber, ceramic, and steel fin burners. The results showed that CO emission decreased and thermal efficiency increased as porosity increased. Khanna et al. [3] designed a burner which consisted of two sections of porous ceramic matrix cylinders in an insulated tube. Result indicated that CO and NO<sub>x</sub> emissions increased with increasing flame speed for a given equivalence ratio, however, NO<sub>x</sub> emissions were fairly constant over the range of the studied flame speed. Mital et al. [4] measured the temperature distributions, stability limits, and pollutants emissions within the submerged reaction zone stabilized inside radiant burners. Results illustrated that CO and HC emissions decreased with a decrease in equivalence ratio and an increase in firing rate. The local temperature and species distributions showed that the structure of submerged flames involved a broad reaction zone and the temperature distributions revealed that the reaction zone moved upstream as the firing rate increased. Bubnovich et al. [5,6] experimentally and numerically studied lean methane-air mixtures combustion in a porous media combustor formed by two beds of different sizes of alumina balls. Results concluded that the pollutants emissions increased and the stable combustion regions enlarged with increase in equivalence ratio. Liu et al. [7] numerically studied the combustion of methane/air mixtures in a two-section porous burner. Results showed that the contour of temperature and velocity changed considerably at the interface of the porous media and near the wall and the temperature distributions varied considerably with changes of the radiative extinction coefficient of the large-pore porous media. In addition, our research group's previous studies by Gao et al. [8–12] mainly investigated the gas fuels (e.g., methane, hydrogen, and propane) combustion in different two-section inert porous media burners, such as porous foams, pileup-pellets beads, and honeycombs. Results indicated that all the fuel types, porous media morphologies, and porous media materials can influence the combustion performances.

Inert porous media combustion can only enhance heat recirculation, improve thermal capacity, and intensify gas mixing or diffusion. However, the inert porous media itself cannot promote the fuel combustion or reduce the pollutants emissions. Inert porous media combustion technology has certain limitations regarding fundamentally reducing the fuel activation energy and the flame temperature. Catalytic combustion is a modified technology for lean methane/air premixed combustion that can overcome the above limitations for the inert porous media combustion. Comprehensive studies have been dedicated to further understanding the catalytic combustion. These studies mainly focused on catalysts, supports, and combustion performances optimizations.

A suitable catalyst plays a vital role in stable and effective catalytic combustion. Supported noble metal oxides possess excellent performance and are most widely investigated by researchers, whereas noble metals are expensive and prone to deactivation due to sintering, decomposition, and undesirable interactions with supports in combustion at elevated temperatures [13]. Therefore, a variety of inexpensive transition metal oxide catalysts, such as perovskites [14] and hexaaluminates [15], have been recently explored. Among the transition metal oxide catalysts, perovskites are one of the most active and stable catalysts at elevated temperature combustion. The activity of perovskites is slightly weaker than that of the noble metal catalysts, but the thermal stability of perovskites and their anti-sintering properties are significantly superior to those of the noble metal catalysts and hexaaluminates [16]. Significant attention has been paid to perovskite-type oxides (general formula ABO<sub>3</sub>) because of their prominent anti-sintering and thermal stability. Fabbrini et al. [17] claimed that the catalytic

activity of perovskites is mainly determined by the B-position element. The substitution of the A-position element with another element possessing a different valence can increase catalytic activity and thermal stability in methane catalytic combustion. Batis et al. [18] reported that lanthanum-based perovskites containing manganese, cobalt, and iron are the most active in methane combustion. Alifanti et al. [19] reported the effect of Ce substitution on the activity of LaMnO<sub>3</sub> perovskites in methane combustion, and proposed that Ce substitution significantly affects the thermal stability and modifies the oxygen desorption characteristics.

The supports play significant roles in maintaining the activity and long-term thermal stability of the catalysts. Several main types of supports, such as monoliths, ceramic foams, and pileup-pellets, are used in methane catalytic combustion at elevated temperatures. Monoliths are mainly used for the supports because of their distinct advantages, such as better mechanical strength and lower thermal expansion coefficients [20]. However, the monolithic catalysts possess separate longitudinal parallel channels resulting in poor heat and mass transfer performances, higher pressure drops, and lower specific surface areas. Foam supports are also commonly used catalysts supports because of their advantages, such as improved gas mixing and preheating, low pressure drop, high porosity, and superior heat and mass transfer coefficients. Nevertheless, foam supports are easily fragile and undergo significant thermal expansion when exposed to elevated temperature and serious thermal shock [21]. The pileup-pellet supports with discrete particles are preferred as alternatives to catalytic combustion at elevated temperatures because of the smaller size and stronger robustness of the particles compared with a rigid and continuous structure. The main advantages of the pileup-pellet supports are significantly improved durability, high heat capacity, and enhanced thermal conductivity. Moreover, the pileup-pellet supports are inexpensive and easy to prepare. Al-attab et al. [22] experimentally investigated low heating value biogas combustion in a packed bed porous media burner with submerged flame. Results concluded that lean stable combustion was achieved with equivalence ratios varied in the range from 0.33 to 0.71 because of the high heat capacity and thermal conductivity for the packed beds; furthermore, both the equivalence ratio and the height of the packed beds can affect the maximum temperatures and the pollutants emissions of the packed bed burner.

The applications of catalytic combustion have been investigated in recent years. Xu et al. [23] investigated premixed catalytic combustion emissions for different air preheating temperatures in catalytic monoliths with ultra-low concentrations of palladium. The results revealed that preheated air generated uniform radial temperatures in catalytic monolith. Thus, preheated air helped to prevent local hot spots and thereby decreased NO<sub>x</sub> emissions. Chen et al. [24] reported that the double perovskite oxide Sr<sub>2</sub>Mg<sub>1-x</sub>Mn<sub>x</sub>MoO<sub>6</sub> ( $x = 0, 0.2, 0.5, 0.8, \text{ and } 1$ ) was used as a catalyst for methane combustion. The results showed that the catalytic performance could be directly correlated with surface Mn composition because the Mn composition prompted the production of the surface lattice oxygen species. Robayoa et al. [25] studied the flammability limits of methane combustion in La-Sr-Fe-Cr-Ru-based perovskite catalysts supported on silicon carbide (SiC) ceramics. The results revealed that coating the SiC porous media with perovskite catalysts can reduce the minimum stable equivalence ratio, increase the combustion reaction rate, and achieve higher combustion efficiency. Banús et al. [26] reported that the structured catalyst composed of a Co, Ba, and K/ZrO<sub>2</sub> layer coated onto metallic foam was a promising system to be used for soot combustion because of its high activity and durability.

Based on the above reviews, the published studies of [1–12] separately focused on the inert porous media combustion. In addition, most of the previous studies of [13–26] on catalytic

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