Fuel 217 (2018) 45–50

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Methane partial oxidation in a two-layer porous media burner with Al_2O_3 pellets of different diameters

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ARTICLE INFO

Keywords:
Partial oxidation
Porous media burner
Pellet diameter
Fuel-rich combustion
Hydrogen

ABSTRACT

In this study, the fuel-rich combustion of methane in a two-layer porous media burner with Al_2O_3 pellets of different diameters was experimentally investigated. The upstream layer consisted of 2.5-mm diameter packed Al_2O_3 pellets, and the downstream layer consisted of 5-mm, 6.5-mm, 7.5-mm, and 9.5-mm diameter packed Al_2O_3 pellets. The effects of pellet diameter on the temperature distribution, exhaust composition, and the syngas energy conversion efficiencies were studied at a fixed operation condition with an equivalence ratio of 1.6 and a gas velocity of 0.13 m/s. An optimal downstream pellet diameter of 7.5 mm was determined for the partial oxidation of methane corresponding to the highest syngas energy conversion efficiency. Stabilized methane fuel-rich flames were realized in the optimized burner for various gas velocities (0.09 m/s–0.15 m/s) and equivalence ratios (1.2–1.7). The effects of operation conditions (gas velocities and equivalence ratios) on the combustion performance were also tested. We found that 50.0% of the methane was converted to H_2 and CO at an equivalence ratio of 1.7 and an inlet gas velocity of 0.15 m/s with burner energy conversion efficiency based on lower heating values.

1. Introduction

Micro combined heat and power (CHP) systems based on solid oxide fuel cells (SOFCs) have been rapidly developed due to their high efficiencies and low emissions. Natural gases are normally fed directly into such systems and are converted to syngas/hydrogen in a fuel processing subsystem through various reforming processes. Steam reforming is currently the most widely used method for the reforming of natural gas [1,2]. However, steam reforming is a very endothermic process that requires an external heat source and additional steam feeds. These additional requirements further increase the system complexity and make steam reforming less suitable for small-scale applications. versus steam reforming, the partial oxidation (POX) of methane is exothermic. It offers simplicity, quick start-up, fast dynamic response and no need for external heat or steam. Therefore, the partial oxidation process has been receiving significant interests from micro CHP systems. The partial oxidation of methane commonly occurs via the aid of a catalyst [3]. However, the catalyst is prone to coking that can easily lead to deactivation. An alternative catalyst-free partial oxidation process is fuel-rich combustion in porous media burners.

Combustion in a porous media burner transfers energy from the flame to the inlet gases through the radiation and heat conductivity of porous solids. This leads to preheating of the reactants, which increases the flame stabilities. There are many studies on fuel-rich combustion in porous media burners. These can be classified into two categories [4]. One is filtration combustion, where transient combustion zones propagate within the porous media [5]. As the flame propagates downstream, the inlet premixed gases adsorb the heat released by the flame and thus are preheated. Until now, most studies have focused on the partial oxidation of methane through filtration combustion in a single-layer of porous media. For example, Bingue et al. studied the ultra-rich filtration combustion of methane formed in an inert porous media for the conversion of methane to syngas [6]. Toledo et al. studied the filtration combustion of methane for various equivalence ratios from 1 to 3.8 and found that 62% of the methane was converted to syngas [7]. Filtration combustion in a porous media burner has also been applied to other hydrocarbons [8]. Toledo et al. studied the syngas production of ethane [9], propane [9–11], butane [11], methanol [12], ethanol [12], diesel fuel [11], and heavy fuel oil [11] via filtration fuel-rich combustion. Ellzey and colleagues studied the filtration fuel-rich combustion of heptane [13], ethanol [14], jet fuel, and butanol [15] for the production of hydrogen and syngas. Meanwhile, several numerical studies on fuel-rich filtration combustion were performed for reactor optimization and mechanistic studies [16–19].

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https://doi.org/10.1016/j.fuel.2017.12.088
Received 12 September 2017; Received in revised form 6 December 2017; Accepted 20 December 2017

Although a high efficiency of 73% can be reached when using methane as the fuel [20], porous media is fragile due to frequent thermal cycles in the filtration combustion. To prevent flame propagation, stationary combustion in which the flames are stabilized in a fixed region has emerged as a safe and feasible method due to material requirements and reliability [21]. In stationary combustion, a two-layer porous media burner supports a stationary combustion zone near the interface of the two layers [22–24]. An upstream porous media layer with a smaller porosity acts as a reactant preheat zone. The downstream layer with a larger porosity acts as a reaction zone for the combustion process. The fuel-lean combustion of methane in a two-layer porous media burnerhas been extensively studied in previous studies [25–28]. Qu and co-researchersstudied the impacts of burner structure and porous media material on combustioncharacteristics [29–31] and introduced catalyst to reduce pollutant emissions [32–34]. While studies on fuel-lean combustion in two-layer porous media burners are plentiful, studies on fuel-rich combustion are rare. Mastorakos and co-researchers studied the stabilized fuel-rich combustion of methane over a range of equivalence ratios. Up to 45% of methane was converted to syngas at an equivalence ratio of 1.85 [35]. Syngas production from liquid fuels was further studied in their subsequent research [36,37]. Trimis and co-workers utilized a two-layer porous media burner for the partial oxidation of methane [38]. Further, they developed a porous media burner with a conical section and a cylindrical section for the stationary fuel-rich combustion of methane [39,40]. In previous studies, we studied the partial oxidation of methane and biogas in two-layer porous media burners [41,42]. The porous media burner developed was then used to supply syngas for the SOFCs in subsequent studies [43–45]. Both ceramic foams and packed Al_2O_3 pellets were used as porous media in the studies. However, the current studies mainly focus on the fuel-rich combustion in packed Al_2O_3 beds with fixed diameters or ceramic foams with fixed porosities. One recent theoretical study evaluated the effects of pellet diameter on the fuel-rich combustion in a two-layer porous media burner [41]. The model showed that the Al_2O_3 pellet diameter had large impact on flame temperature and exhaust compositions. Although simulation results have provided a theoretical base for the prediction of combustion characterizations, experimental data are still critical to validate the model and optimize burner structures.

Thus, we designed and built a two-layer porous media burner with 2.5-mm Al_2O_3 pellets in the upstream section and Al_2O_3 pellets of different diameters (5 mm, 6.5 mm, 7.5 mm, and 9.5 mm) in the downstream section. The effects of the Al_2O_3 pellet diameter on the temperature distribution, flame compositions, and conversion of methane to syngas were examined to identify an optimal burner structure for partial oxidation of methane. The characterization of methane fuel-rich combustion in the optimized porous media burner was tested at various gas velocities and equivalences.

2. Experimental section

2.1. Experimental setup

Fig. 1 shows the schematic of the experimental system. This consists of a gas supply system, a two-layer porous media burner, and a measurement system. Methane and air were regulated using mass flow controllers and then mixed in the premix chamber of the burner. The methane/air mixture flowed through a flashback protection chamber. This chamber was filled with 1-mm to 2-mm diameter quartz sands to prevent flame flashback. The shell of the burner was a stainless steel tube 54 mm in diameter and 200 mm long. Inside the stainless steel tube, there was a cylinder of aluminum silicate arranged for heat insulation. The internal diameter of the cylinder was 30 mm, and the external diameter was 54 mm. In addition, the burner outside was surrounded with a quartz fiber 65 mm thick to ensure that the outside of the insulation was near room temperature. The porous media burner consists of two layers of packed Al_2O_3 pellets. The upstream layer was filled with 2.5-mm Al_2O_3 pellets that were 20 mm long. This acted as a preheat zone for the methane/air mixture. The downstream layer was the flame region and comprised a 60-mm-long packed bed of Al_2O_3 pellets of different diameters (5 mm, 6.5 mm, 7.5 mm, and 9 mm).

Seven S-type thermocouples were spaced 10 mm apart from each other and were labeled as T1, T2, T3, T4, T5, T6, and T7 from upstream to downstream. The temperature data was sampled and recorded via a temperature-sampling module connected to a computer. The combustion exhausts at the burner outlet were sampled by a sampling probe and were measured by the gas chromatograph (GC) with a thermal conductivity detector (TCD). The GC conditions are shown in Table 1. The measured CO composition was revised by conducting a mass balance check process because there was a big overlap between the CO peak and N_2 peak in the GC measurement. This was explained in detail in a prior study [41].

2.2. Parameter definitions

The inlet velocity is calculated by

$$v = \frac{\dot{V}_{\text{in}}}{A} = \frac{4\dot{V}_{\text{in}}}{\pi D^2} = \frac{4(\dot{V}_{\text{CH}_4} + \dot{V}_{\text{air}})}{\pi D^2} \tag{1}$$

where $\dot{V}_{\text{in}}$ m^3/s is the gas flow rate of methane/air mixture, A (m^2) is the sectional area of the flow path, D (m) is the diameter of the flow path, $\dot{V}_{\text{CH}_4}$ m^3/s and $\dot{V}_{\text{air}}$ m^3/s are the gas flow rates of methane and air respectively.

The equivalence ratio is defined as

$$\phi = \frac{(m_{\text{fu}}/m_{\text{air}})_{\text{act}}}{(m_{\text{fu}}/m_{\text{air}})_{\text{stoic}}} = 9.52 \times \frac{\dot{V}_{\text{CH}_4}}{\dot{V}_{\text{air}}} \tag{2}$$

where $(m_{\text{fu}}/m_{\text{air}})_{\text{act}}$ is the actual ratio of the mass flow rate of fuel and air, and $(m_{\text{fu}}/m_{\text{air}})_{\text{stoic}}$ is the stoichiometric ratio of the mass flow rate of fuel and air.

The syngas energy conversion efficiency was defined to represent the amount of the chemical energy of syngas in the product species compared to that of the inlet fuel [5,13]:

$$\eta_{\text{e-s}} = \frac{y_{\text{H}_2} \times \text{LHV}_{\text{H}_2} + y_{\text{CO}} \times \text{LHV}_{\text{CO}}}{y_{\text{CH}_4,\text{in}} \times \text{LHV}_{\text{CH}_4}} \tag{3}$$

where y_{H_2} and y_{CO} are the mass fractions of H_2 and CO in the combustion exhausts respectively, $y_{\text{CH}_4,\text{in}}$ is the mass fraction of CH_4 in the inlet mixture, $\text{LHV}_{\text{H}_2} = 120.1$ MJ/kg,

$\text{LHV}_{\text{CO}} = 10.1$ MJ/kg, and $\text{LHV}_{\text{CH}_4} = 50.2$ MJ/kg are the lower heating values of H2, CO and CH4, respectively.

Except for the syngas energy conversion efficiency, the conversion of methane was also defined to quantify the extent of the reaction. The conversion of methane can be calculated as

$$\eta_{\text{conv}} = \frac{\dot{m}_{\text{CH}_4,\text{in}} - \dot{m}_{\text{CH}_4,\text{out}}}{\dot{m}_{\text{CH}_4,\text{in}}} = \frac{y_{\text{CH}_4,\text{in}} - y_{\text{CH}_4,\text{out}}}{y_{\text{CH}_4,\text{in}}} \tag{4}$$

where $y_{\text{CH}_4,\text{out}}$ is the mass fraction of CH_4 in the outlet exhausts.

The uncertainty analysis process for the input parameters and the data have been described previously [41], thus not shown in this paper. The calculated experimental uncertainties are shown in Table 2.

2.3. Experimental procedure

First, a premixed methane/air mixture with an equivalence ratio of 0.8 and an inlet velocity of 0.15 m/s was ignited at the burner outlet. Initially, a blue surface flame appeared at the burner outlet. The flame then propagated into the porous media and preheated the burner. When the flame reached the interface of the two layers, i.e., the maximum temperature reached the thermocouple T2, the gas flows were regulated to a fuel-rich condition with an equivalence ratio of 1.6 and an inlet velocity of 0.15 m/s. The heat released by the methane/air mixture