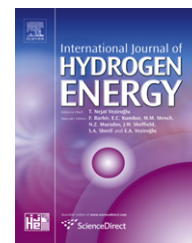


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Design of a novel hydrogen–syngas catalytic mesh combustor

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ABSTRACT

A small-scale wire-mesh catalytic combustor is developed and evaluated for hydrogen–syngas combustion in domestic power/heating generator. The single- and double-layer wire-mesh catalysts are tested to verify their performance on CO conversions. Experimental results indicate that the double-layer wire-mesh catalytic combustor yields a higher CO conversion ratio (>90%) than that (<40%) of the single-layer wire-mesh catalyst in the range of fuel concentrations, fuel compositions, and flow velocities studied. In order to maintain a stable heterogeneous/homogeneous reaction at the second stage of wire-mesh catalyst, a minimum of 4% hydrogen in syngas and at least 200 °C of preheating temperature on the second wire-mesh catalyst are suggested. The advantages of the wire-mesh combustor are its compactness and ease of assembly and cleaning.

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1. Introduction

Modern technique such as integrated gasification combined cycle (IGCC) has enabled combustion of coal, biomass, and other low-grade solid or liquid fuels while still maintaining high conversion efficiencies and low pollutant emissions. Syngas derived from coal and biomass is particularly promising in this regard. The primary constituents of syngas are CO and H₂ with concentration ratios usually in the range of 0.5–2 and it may also contain small amount of N₂, CO₂, H₂O, CH₄, and other higher-order hydrocarbons [1,2]. Besides, syngas is one of the important timely energy resources and sources of massive hydrogen supply. The different CO/H₂ compositions have different flame speeds, flammability limits, and combustion characteristics [3–7]. Hydrogen in syngas plays an important role to improve lean-flame stability and extend flammability limit [8,9]. However, the syngas combustion has its inherent defect of low calorific (BTU) value and low energy density. Thus it yields low flame temperature, narrow flammability limits, and low combustion efficiencies.

Previous investigations have shown that catalytic combustion is a very promising technique to enhance combustion stability in the fuel-lean conditions, to reduce the pollutant emissions, and to extend the flammability limit of the low-BTU flames. These facts suggest that the use of catalysts seems to be very well suited for the low-BTU, low operation temperature syngas-based fuel combustion. Groppi et al. [10] have studied the combustion of syngas/air on the magnesium-substitute hexaaluminate catalysts in a hybrid combustor. Zwinkels et al. [11] and Pocaroba et al. [12] have respectively studied the effect of high-temperature and ageing on Pt- and Pd-based catalysts for biogas combustion. While Johansson et al. [13] reported the development of hexaaluminate catalysts for gasified biomass combustion in gas turbine combustors. These investigations [11–13] have shown that the characteristics of hydrogen reaction on the catalyst surface are completely different from those of the other components in syngas. Hydrogen tends to react and release heat earlier in the catalyst bed, usually within few millimeters, while methane and carbon monoxide are reacted later at

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the tail of the catalyst bed. To design a syngas combustor with a fixed-length catalyst bed, the major concern is the trade-off between fuel conversion ratio and heat loss. The other issue is the reaction of hydrogen in the entrance region of the catalyst bed that may lead to local sintering of the catalyst because of the high-temperature combustion.

Regarding to the configurations of catalyst bed, the wire-mesh catalysts offer the advantages such as high mass and heat transfer performance, low pressure-drop, and ease of assembly and cleaning over the pellet-type and monolith catalysts [14]. Ahlström-Silversand and Odenbrand [15,16] compared the performances of wire-mesh, monolith and pellet catalysts and found that the wire-mesh catalysts possess high mass and heat transfer, moderate pressure-drop, and insignificant effects of pore diffusion as well as axial dispersion. Moreover, the effects of catalyst clogging and fouling can be easily handled with the wire-mesh catalysts. In addition, Jannasch et al. [17] also showed that the wire-mesh catalysts are very promising for natural gas combustion with respect to the catalyst life-time, the NO_x and the CO emissions.

For the design of a catalytic combustor, the operation (sintering) temperature, low-temperature ignition, lightoff temperature, flame stabilization and lean blowout, combustor outlet temperature, and combustion efficiency (or fuel conversion ratio) must be considered. These factors are very important and sometimes conflict with each other. Trade-off and special design considerations are usually applied to optimize the performance of a catalytic combustor. In the present study, the ignition of syngas with minimum energy input and the stabilizing of fuel-lean combustion are the principal considerations of a small-scale, double-layer wire-mesh catalytic combustor design. The design concept is to make use the characteristics of easy lightoff and quick thermal response of the wire-mesh catalysts for hydrogen ignition at the first layer, and then to release heat quickly for preheating and stabilizing the fuel-lean-syngas combustion on the second layer of the wire-mesh catalyst. Therefore, the problems associated with local catalyst sintering, excessive heat loss, and incomplete fuel conversion can be properly alleviated for different operating conditions. In this study, the catalyst configuration, the number of wire-mesh layer, and the separation distance (D) of the wire-mesh are examined and addressed in the following sections.

2. Experiments

Fig. 1 shows the photograph and schematic diagram of the double-layer wire-mesh catalytic combustor. The combustor chamber is made of quartz-tube with an inner diameter of 2.5 cm. The wire-mesh catalysts are clamped between the flange and the quartz-tube. Note that the separation distance between the wire-mesh catalysts is 1–3 cm. The larger distance appeared in the photograph is to show the wire-mesh clearly. The commercial catalyst is made of metal alloy and coated with platinum. The blockage ratio, which is defined as the ratio of the metal area over the total area of the wire-mesh, is 48%. Air is supplied from a compressor system whereas hydrogen and carbon monoxide with various

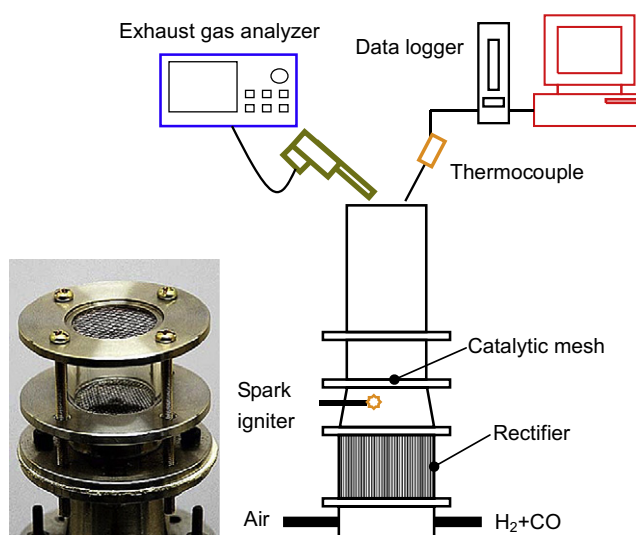


Fig. 1 – Photograph and schematic diagram of the double-layer wire-mesh catalytic combustor.

volumetric ratios are supplied from gas cylinders to simulate syngas. Fuel and air are premixed and rectified before entering the combustor. A commercial spark igniter with minimum energy input is used for the cold start in the combustor. Gas emissions are measured by a gas analyzer (MRU) and a gas chromatography (Varian). Concentration of CO is calibrated in the range of 0–10,000 ppm with an uncertainty of ± 20 ppm. The uncertainty of hydrogen measured by the gas chromatography is within 2%. A K-type thermocouple is used to measure the temperature with a BeO and 10–15% Y_2O_3 coating to eliminate catalytic reaction.

3. Results and discussion

3.1. Ignition process

Preliminary tests indicate that the 6% of H_2 in the syngas can be easily ignited by the spark igniter and produce a stable flame, as shown in Fig. 2(a). The flame is then to quickly heat up the wire-mesh catalysts and to stabilize the catalytic reaction in the combustor even when the spark igniter is turned off and removed, as shown in Fig. 2(b). When the catalytic reaction of syngas undergoes, the volumetric percentage of H_2 in the syngas is adjusted to the prescribed experimental conditions. It is noted that the use of a commercial spark for the cold start is more reliable and controllable than employing the method of auto-ignition of catalyst at low-temperature.

For the catalytic reaction of syngas, hydrogen is reacted on the first-layer wire-mesh catalyst to release sufficient heat for preheating the unreacted species and the second-layer wire-mesh catalyst. The preheated flue gas from the first stage contains chemical radicals that could sustain a homogenous reaction downstream and stabilize the lean-syngas catalytic combustion [18,19].

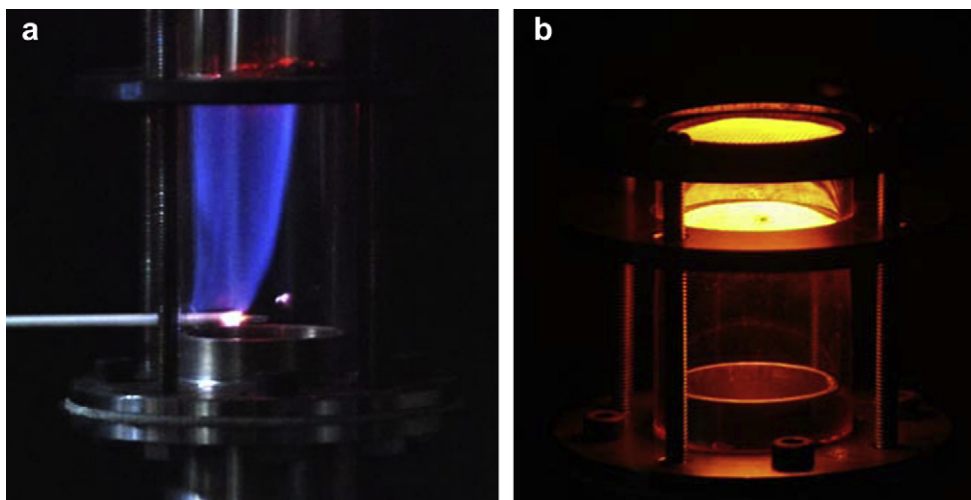


Fig. 2 – Ignition process of the double-layer wire-mesh catalytic combustor (a) during ignition and (b) after ignition.

3.2. Catalyst configuration and arrangement

In order to examine the feasibility of the wire-mesh catalytic combustor, three different catalyst configurations are examined. Fig. 3 compares the CO conversion ratio of the premixed syngas ($5\%H_2 + 5\%CO$, $\phi = 0.264$) reaction on the honeycomb, single-layer, and double-layer wire-mesh catalysts. According to the small-scale power/heating generator which is supposed to produce 1–10 kW [20], its corresponding velocities range from 1 to 9 m/s based on $5\%H_2 + 5\%CO$ fuel concentration. The double-layer wire-mesh catalyst results in the highest CO conversion ratio (>90%), whereas the single-layer wire-mesh catalyst yields the lowest CO conversion ratio (<40%) in the range of flow velocities studied. The honeycomb catalyst has the relatively high CO conversion ratio (>85%) in high flow velocity regions. In principle, the monolith honeycomb needs more thermal input to heat up and maintain the catalyst

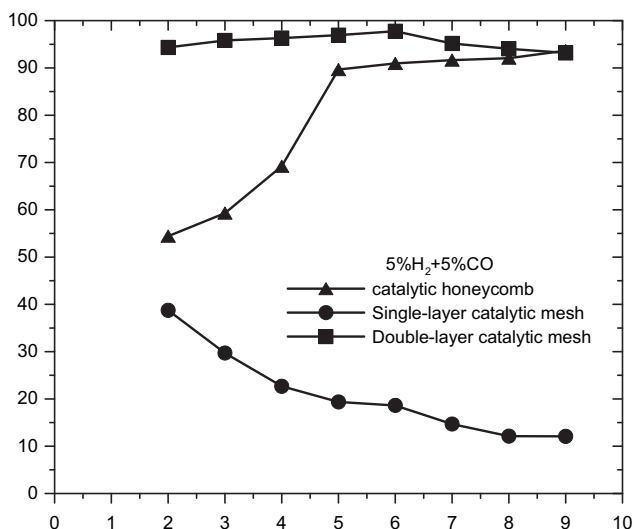


Fig. 3 – CO conversion ratios measured from three different catalyst configurations.

temperature than that required by the wire-mesh catalysts. Therefore, it takes more fuel–air mixture per unit time, in terms of high flow velocity as shown in the figure, to have high CO conversion. It is noted that the CO conversion ratio of the single-layer wire-mesh catalyst decreases with increasing the flow velocity. As the flow velocity is increased, the residence time on the catalyst is shortened, and the cooling of the catalyst is also increased. Thus the CO cannot be effectively converted to CO_2 by the single-layer wire-mesh catalyst. However, when a second wire-mesh catalyst is added to the downstream of the first one, the CO conversion ratio is increased substantially. The idea of adding the second catalyst to the downstream of the first one is to continue the heterogeneous/homogeneous reaction. Hydrogen in syngas is supposed to react and release energy earlier in the first catalyst than the CO, due to its higher diffusivity and reactivity. High-temperature product gas with the unburned mixture from the first catalyst is then to heat up and trigger the catalytic reaction on the second catalyst for the rest of CO reaction. Accordingly, the double-layer wire-mesh catalyst configuration has an excellent CO conversion ratio in both low and high flow velocities as compared to the single-layer wire-mesh and the honeycomb catalysts.

3.3. Separation distance between two wire-meshes

Although the double-layer wire-mesh catalyst inherits both benefits of fast heat transfer of the metallic mesh and high thermal energy preservation of the monolith honeycomb, the separation distance (D) between the two meshes is an essential parameter to ensure the high performance of the catalytic combustor. Too large a separation distance may lead to an increase of heat loss of the gas, but too short a distance may curtail the residence time of the gas within the catalytic meshes. In order to determine the optimum separation distance for the combustor design, several cases such as $D = 0.5, 1, 2,$ and 3 cm are tested to examine the performance of CO conversion. For $D = 0.5$ cm, the short separation distance of the wire-mesh results in a poor performance of CO

conversion. While the CO conversion obtained from $D = 2$ cm is in between that from $D = 1$ and 3 cm. Therefore, we only present experimental results obtained from $D = 1$ and 3 cm for further discussion. Fig. 4 compares the CO conversion ratios measured from two different separation distances ($D = 1$ and 3 cm) and syngas mixtures ($4\%H_2 + 4\%CO$, $\phi = 0.207$ and $5\%H_2 + 5\%CO$, $\phi = 0.264$). It can be seen that the CO conversion ratios for $D = 1$ cm are higher than those for $D = 3$ cm for both syngas mixtures. This fact suggests that $D = 1$ cm is an optimum separation distance for the present combustor design under the various syngas mixtures and flow velocities studied.

In order to understand the effect of gas temperature on the performance of the wire-mesh catalysts, the temperatures just below and above the second wire-mesh catalyst are measured and shown in Fig. 5. The temperature measured just below the second catalyst is to monitor the thermal condition of the remaining syngas incoming to the catalyst. The sufficiently high-temperature of the remaining syngas is a vital key to sustain chemical reaction at the second catalyst. On the other hand, the temperature measured above the second catalyst is to examine if the chemical reaction is completed. Fig. 5 shows that the temperatures measured at the locations below and above the catalyst are slightly higher for $D = 1$ cm than those for $D = 3$ cm. And the higher H_2 contents of the syngas produce the higher temperatures. The decrease of the incoming gas temperature to the second catalyst with increasing the flow velocity is due to the reduced residence time that leads to incomplete reactions of H_2 and CO on the first catalyst. The unreacted H_2 and CO are then convected to the downstream, react at the second catalyst, and release more energy to increase the exhaust gas temperatures in the higher flow velocity regions (>5 m/s).

3.4. Effect of Syngas Concentration

It has been shown that the double-layer wire-mesh catalytic combustor with $D = 1$ cm has an excellent performance on CO conversion. However, the concentrations of the syngas may also influence the performance of the combustor. To study the

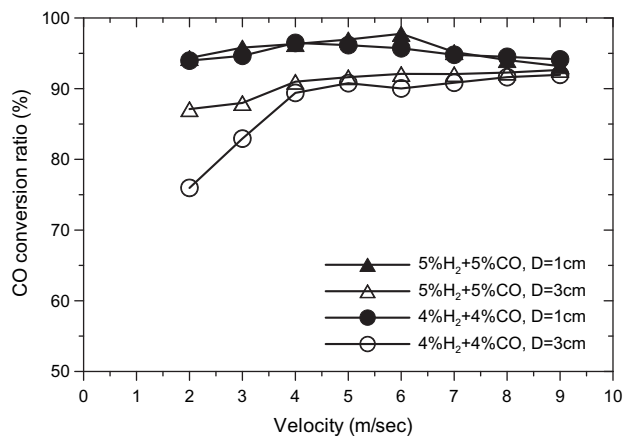


Fig. 4 – CO conversion ratios measured in the double-layer wire-mesh catalytic combustor with $D = 1$ and 3 cm under various fuel concentrations and flow velocities.

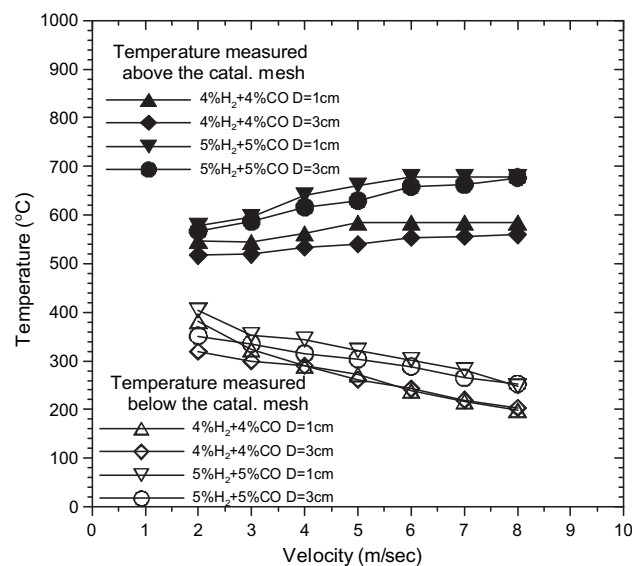


Fig. 5 – Temperatures measured at the locations just below and above the second catalyst in the double-layer wire-mesh catalytic combustor with $D = 1$ and 3 cm under various fuel concentrations and flow velocities.

effects of syngas concentrations on fuel conversions, three different syngas mixtures ($3\%H_2 + 3\%CO$, $4\%H_2 + 4\%CO$ and $5\%H_2 + 5\%CO$) are employed. Fig. 6 shows the H_2 and CO conversion ratios on the single-layer wire-mesh catalyst. It can be seen that the conversion ratios of H_2 are higher than those of CO in all flow velocities. This fact, again, indicates that the H_2 reacts faster than the CO on the catalyst. The concentration of $3\%H_2 + 3\%CO$ in the syngas yields the highest H_2 and lowest CO conversion ratios than the other two

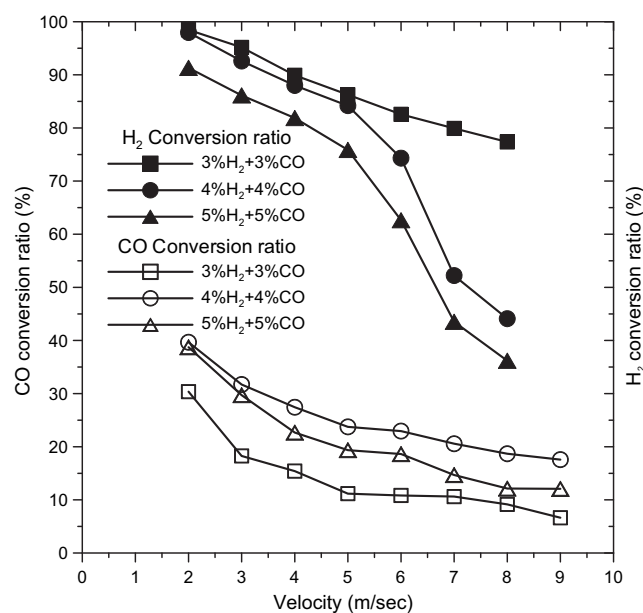


Fig. 6 – H_2 and CO conversion ratios measured in the single-layer wire-mesh catalytic combustor under various fuel concentrations and flow velocities.

cases. When the flow velocity is increased to 6 m/s, the H_2 conversion ratios decrease to less than 80% for the concentrations of $4\%H_2 + 4\%CO$ and $5\%H_2 + 5\%CO$ due to the reduced residence time. This leads to at least 1% of the unreacted H_2 remained in the gas stream. These unreacted H_2 concentrations for the cases of $4\%H_2 + 4\%CO$ and $5\%H_2 + 5\%CO$ will improve the CO conversions when the second wire-mesh catalyst is added to the combustor.

Fig. 7 compares the CO conversion ratios on the single- and double-layer wire-mesh catalysts for three different syngas concentrations. The double-layer catalyst with $D = 1$ cm has excellent CO conversion ratios (>90%) for the $4\%H_2 + 4\%CO$ and $5\%H_2 + 5\%CO$ concentrations, but is not for the case of $3\%H_2 + 3\%CO$ (<60%). In addition, the double-layer catalyst generates higher CO conversions than the single-layer catalyst for various fuel concentrations and flow velocities. In order to understand why the CO conversions for the case of $3\%H_2 + 3\%CO$ are much lower than those for the $4\%H_2 + 4\%CO$ and $5\%H_2 + 5\%CO$ concentrations, temperature measurements are performed at the locations just below and above the second catalyst. Fig. 8 shows that the temperatures measured at both locations are all lower for the case of $3\%H_2 + 3\%CO$ than those for the $4\%H_2 + 4\%CO$ and $5\%H_2 + 5\%CO$ conditions. The measured temperature ahead of the second catalyst is about $270^\circ C$ at low flow velocity (2 m/s) and decreases progressively to $130^\circ C$ when the flow velocity reaches to 8 m/s. Therefore, it is conjectured that the 3% of H_2 in the syngas releases insufficient heat for preheating the second catalyst and assisting further CO reactions. Besides, the $3\%H_2 + 3\%CO$ concentration in the syngas mixture ($\phi = 0.152$) is almost out of the lean flammability limit (4 vol.% for H_2 and 21 vol.% for CO). The remaining H_2 concentration for the $3\%H_2 + 3\%CO$ condition after passing the first catalyst is less than 1% due to its high H_2 conversion ratio as presented in Fig. 6. These two

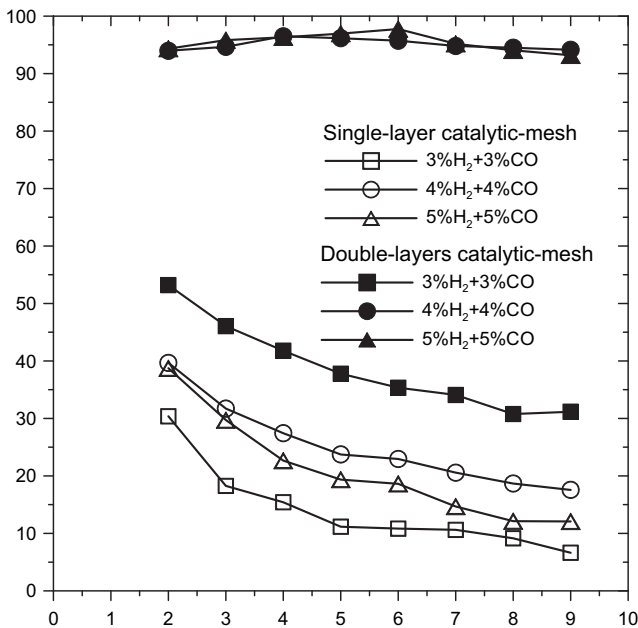


Fig. 7 – CO conversion ratios measured in the single- and double-layer ($D = 1$ cm) wire-mesh catalytic combustor under various fuel concentrations and flow velocities.

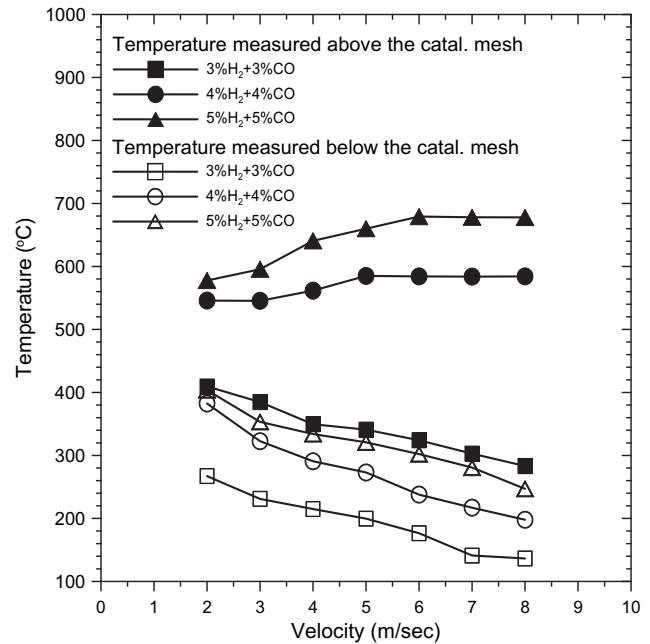


Fig. 8 – Temperatures measured at the locations just below and above the second catalyst in the double-layer wire-mesh catalytic combustor with $D = 1$ cm under various fuel concentrations and flow velocities.

factors, low preheating temperature and less H_2 concentration, may lead to the much lower CO conversions for the case of $3\%H_2 + 3\%CO$. Low CO conversion ratio is unacceptable for the combustor design. Therefore, in order to preserve high CO conversion ratio in the double-layer wire-mesh catalytic combustor, a minimum of 4% H_2 concentration in syngas and a preheating temperature of $200^\circ C$ on the second catalyst are suggested.

3.5. Effect of syngas composition

In general, the fuel composition ratio of CO to H_2 in syngas is usually in the range of 0.5–2. In order to study the effect of fuel composition on the CO conversions, three different fuel

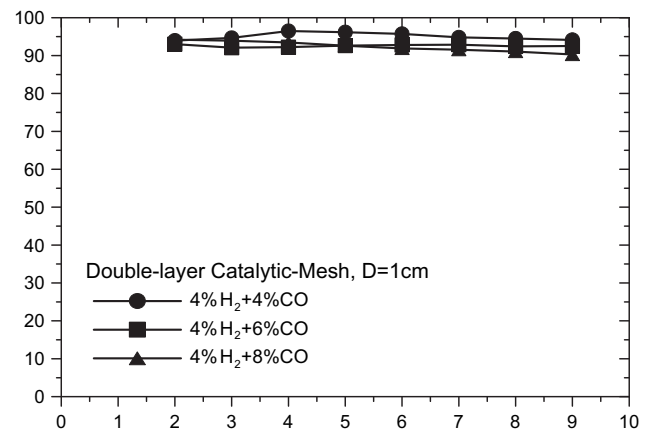


Fig. 9 – CO conversion ratios measured in the double-layer wire-mesh catalytic combustor with $D = 1$ cm under various fuel compositions and flow velocities.

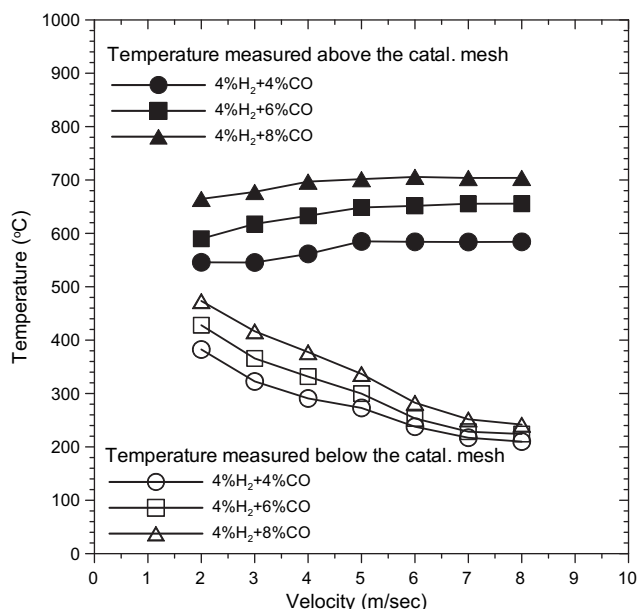


Fig. 10 – Temperatures measured at the locations just below and above the second catalyst in the double-layer wire-mesh catalytic combustor with $D = 1$ cm under various fuel compositions and flow velocities.

composition ratios of $\text{CO}/\text{H}_2 = 1, 1.5$ and 2 are tested in the double-layer wire-mesh catalytic combustor. Fig. 9 displays the CO conversion ratios for the fuel composition of $4\% \text{H}_2 + 4\% \text{CO}$ ($\phi = 0.207$), $4\% \text{H}_2 + 6\% \text{CO}$ ($\phi = 0.264$) and $4\% \text{H}_2 + 8\% \text{CO}$ ($\phi = 0.324$). Experimental results indicate that the CO conversion ratios are all higher than 90% and no significant differences among the CO conversions for the three fuel compositions studied. It appears that a 4% of H_2 in syngas is sufficient to support the heterogeneous/homogeneous reaction of CO in the combustor. This verifies that the double-layer wire-mesh catalytic combustor has an excellent performance on CO conversions for various syngas compositions.

Fig. 10 shows the temperatures measured at the locations just below and above the second catalyst for three different fuel compositions and various flow velocities. It can be seen that the gas temperatures measured at both locations increase with increasing CO concentration for all flow velocities. It is believed that part of the CO is reacted on the first catalyst and the reaction is increased with increasing CO concentration leading to a higher gas temperature ahead of the second catalyst. Higher incoming temperatures and higher CO concentrations result in higher product temperatures behind the second catalyst.

4. Conclusions

A small-scale wire-mesh catalytic combustor is developed and evaluated for high efficiency hydrogen-syngas combustion. The single- and double-layer wire-mesh catalysts are tested to verify their performance on CO conversions. A commercial spark igniter with minimum energy input is used for the cold

start in the combustor. Experimental results indicate that the double-layer wire-mesh catalytic combustor with $D = 1$ cm yields a higher CO conversion ratio ($>90\%$) than that ($<40\%$) of the single-layer wire-mesh catalyst in the range of fuel concentrations, fuel compositions, and flow velocities studied. It is found that H_2 in syngas reacts and releases energy earlier in the first catalyst than the CO, due to its higher diffusivity and reactivity. In order to preserve high CO conversion ratio in the double-layer wire-mesh catalytic combustor, a minimum of 4% H_2 concentration in syngas and at least 200°C of preheating temperature on the second wire-mesh catalyst are suggested. Besides, experimental tests also indicate that the double-layer wire-mesh catalytic combustor has an excellent performance on CO conversions for various syngas compositions.

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