Experimental study on premixed methane-air catalytic combustion in rectangular micro channel

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Abstract

The catalytic combustion process of pre-mixed methane-air in a rectangular micro channel was studied experimentally. Infrared thermal imager and flue gas analyzer were used to measure the temperature distribution of the outer wall and main components of exhaust gas respectively. Flammability limits of premixed methane-air in catalytic and non-catalytic micro channel were obtained by changing flow rates of methane, and the effects of equivalence ratio, inlet velocity and channel height on combustion characteristics were analyzed. Results showed that the flammability limits improved significantly when platinum was added into the micro channel. The highest centerline temperature of the outer wall was obtained at an equivalent ratio of 0.9 at the same inlet velocity with or without catalyst in the micro channel. Addition of catalyst in the channel not only gave a uniform temperature distribution on the outer wall of the channel but also improved methane conversion. With the increase of inlet velocity, the centerline temperature of the outer wall increased and the highest points of the temperature shifted to downstream of the channel gradually. Combustion intensity in the channel increased with the increase of channel height at the same inlet velocity.

Key words: rectangular micro channel; micro combustion; catalyst; flammability limit; experimental study

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1. Introduction

From the developmental history of modern society and the evolution of power machinery, the development of industrialization cannot leave the guarantee of stable and continuous power generating facilities. At present, all kinds of micro devices are driven by traditional chemical batteries. Although traditional chemical batteries are simple and reliable, and the technology is mature, its low energy density and longer charging time limits its development. Micro Electro Mechanical System (MEMS) with hydrocarbon fuel has the advantages of high energy density, small size, light weight and low cost and it has wide application prospect. Thus, power MEMS based on combustion has more potential than the conventional chemical batteries. Research works have been carried out by many research institutes from all over the world [1-3]. The core component of power MEMS is the micro combustor, which has a very small combustion space and very short residence time of fuel and oxidizer. Because the characteristic size of micro combustor is close to or less than the quenching diameter or quenching distance, there is a sharp increase in combustion instability [4]. In addition, with the reduction in the size of the combustor, the larger ratio of surface to volume leads to greater heat loss through the wall [5]. Therefore, it is hard to guarantee the complete combustion of fuels, and the blow off limits and ranges of stable combustion are reduced greatly [6], so it is an urgent problem to improve the stability and sufficiency of combustion in the micro combustor.

Many measures have been taken to improve the stability of combustion in micro combustors by scholars from different countries [7-9]. Including the use of special structure design by Wan J et al. [10], Jiaqiang E et al. [11], and Yang W M et al. [12-13], adding porous media in the combustion chamber by Pan J F [14] and Yang W M et al. [15], and using bluff body to improve the stability of combustion by Bagheri G et al. [16]. Besides, catalytic combustion is also an effective and simple method to guarantee stable combustion [17-20]. The reason lies in that, the reduction in the size of combustor will result in insufficient and unstable combustion. But the ratio of surface-to-volume will increase, and the relative increase in the surface area of inner face can provide a favorable condition for the surface catalytic reaction. However, the internal combustion process will become complex because of the existence of surface reaction. Numerical simulation and experimental research have been used to research the combustion characteristics of different fuels and oxidizers in micro combustors. Chen et al. [21] performed numerical simulations to analyze the reactions of hydrogen/air inside a catalytic micro-tube with detailed heterogeneous and homogeneous chemistries. They studied the characteristics of heterogeneous and homogeneous interaction in terms of flow velocity, tube diameter and wall thermal conductivity. Benedetto et al. [22] conducted two-dimensional CFD simulations to investigate the catalytic micro-combustor and the non-catalytic micro-combustor. Their numerical results showed that the catalyst coated micro combustor can be operated at high inlet gas velocities and input high powers. Kamada T et al. [23] investigated the combustion and ignition characteristics of natural gas components in a micro flow reactor with a controlled temperature profile experimentally and computationally. Their results indicated a significant effect of n-butane addition in the blends on combustion and ignition characteristics of the blended fuels. Davis et al. [24] studied the contributions of homogeneous and heterogeneous reactions to high-temperature catalytic methane oxidation over three different gauze catalysts (Pt, Pt-10%Rh, and Ni) using laser-induced fluorescence (LIF) spectroscopy. They found that Pt is the most active oxidation catalyst among the three catalysts and with the highest catalytic activity. Maruta et al. [25] computationally studied the extinction limits of methane-air mixtures in a micro scale tube reactor coated with Pt catalyst. They found that the exhaust-gas recirculation rather than lean mixtures are preferable for minimizing flame temperatures in catalytic micro combustors. To enrich the basic research data for micro combustion, and obtain the impact of relevant factors on combustion process, the flammability ranges of premixed methane-air in micro channel were tested in this paper, and the effect of some vital parameters on the stable combustion characteristics were researched, such as equivalence ratio, inlet velocity and the channel height.

2. Experimental setup and method

Schematic diagram of the experimental set-up is shown in Fig. 1. The whole experimental system includes a fuel and oxidizer supply system, a control system and an observation and recording system. The fuel and oxidizer were released by high pressure gas cylinders into the premixed chamber through the flow control system for sufficient mixing, and eventually ignited in the micro channel. The observation and recording system includes an infrared thermal imager, a gas analyzer and other auxiliary equipment.

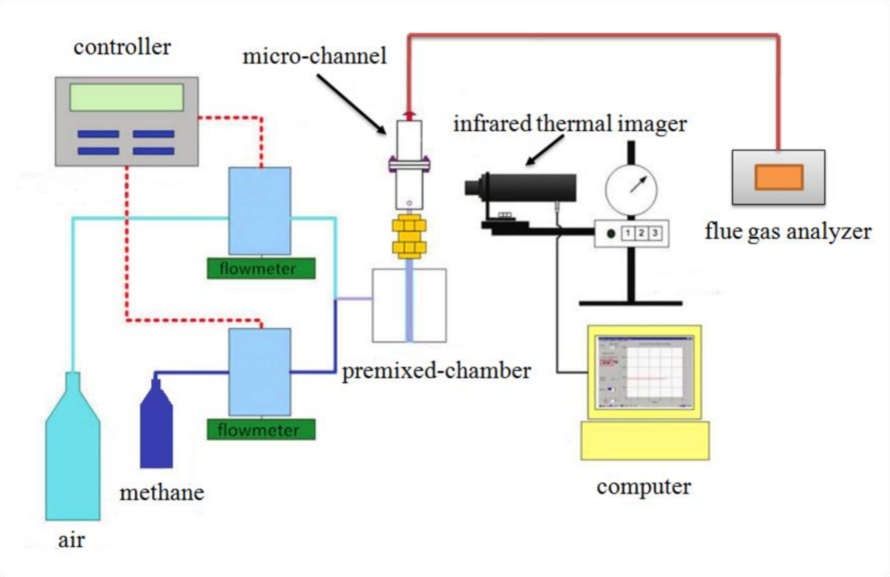

Fig. 1. Schematic diagram of experimental system.

The structure of rectangular micro channel used in the experiment is shown in Fig. 2. It was made of 316L stainless steel, and three kinds of channel height were designed. The internal sizes of the micro channel are 20 mm × 10 mm × 3 mm, 20 mm × 10 mm × 2.5 mm and 20 mm × 10 mm × 2 mm respectively. The catalyst is platinum, and the size of the catalytic surface is 20 mm × 10 mm. The fuel and oxidizer for the experiment were methane and air respectively. Methane has a purity of more than 99.9% and the outlet pressure for both fuel and oxidizer was 0.15 Mpa. The ambient temperature is 293 K and the environmental relative humidity is within 76%. The inlet velocity and equivalence ratio of methane and air were set by the mass flow controller, which is produced by MKS Company of America and it can be used in many situations to measure and control the gas flow rate, especially when the situation required a high repeatability. Its accuracy is up to ±1 % and has a response time less than 1 second. The model of the infrared thermal imager is Thermovision A40, and its minimum focal length is 4 mm. It can detect the temperature change between -40 to 2000 ℃ and its measurement accuracy is ±2 %. In the experiment, the VARIO PLUS flue gas analyzer was used, and its measurement accuracy was up to ±0.2 %, which can be used to analyze the exhaust gas for a long time continuously.

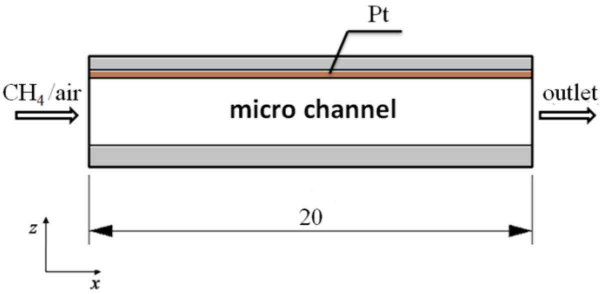

Fig. 2. Section structure of catalytic micro channel.

3. Results and Discussions

3.1. Analysis of flammability limits

In order to analyze the flammability limits of the micro channel and the ranges of stable combustion, keeps the flow rate of methane constant at first, then increasing the flow rate of air, the flame will stabilize in the channel, and this equivalence ratio is the flammability limits under fuel rich condition. After the flame has stabilized in the channel, increase the flow rate of air gradually until the flame is blown out of the channel, and this equivalence ratio is the flammability limits under oxygen rich condition. Then, changing the flow rate of methane and repeating the above experimental process, the flammability limits and ranges of stable combustion in the micro channel under different methane flow rates are obtained. The height of the micro channel is 3 mm, and the methane flow rate is increased from 50 sccm to 160 sccm. Fig. 3 is the flammability limits of catalytic and non-catalytic micro channel under different flow rates of methane. It can be seen that with the increase of the flow rate of methane, the flammability limits under fuel rich conditions increase at first and then decrease. When the flow rate of methane is over 80 sccm, the drop slope of flammability limits under fuel rich conditions increases gradually with the increase of the flow rate of methane for the two kinds of micro channels. The flammability limits under oxygen rich conditions decrease linearly with the increase of the flow rate of methane, but the reduction of the flammability limits under oxygen rich conditions in the catalytic micro channel is less than that of the non-catalytic micro channel. For the two kinds of micro channels, with the increase of the flow rate of methane, the ranges of stable combustion all decrease gradually under oxygen rich conditions, but its range in catalytic channel is significantly larger than that of the non-catalytic channel. For instance, when the flow rate of methane is 100 sccm, the equivalence ratio of flammability limits in catalytic micro channel are 0.68 and 1.12, which is larger than that of non-catalytic micro channel, in which the flammability limits are 0.71 and 1.08. Thus, we can draw a conclusion that the addition of catalyst in micro channel can improve the stability of combustion greatly. When the flow rate of methane is low, the flammability limits under fuel lean conditions in the catalytic channel are lower than that of the non-catalytic channel, which indicates that the effect of the catalyst is not obvious under the fuel lean and low flow rate conditions. With the increase of the flow rate of methane, the flammability limits under fuel lean conditions in catalytic channel are significantly higher than that of the non-catalytic channel, which indicates that when the flow rate of methane is high, the addition of catalyst in micro channel can strengthen the combustion reaction, and improve the stability of combustion for methane-air in micro channel.

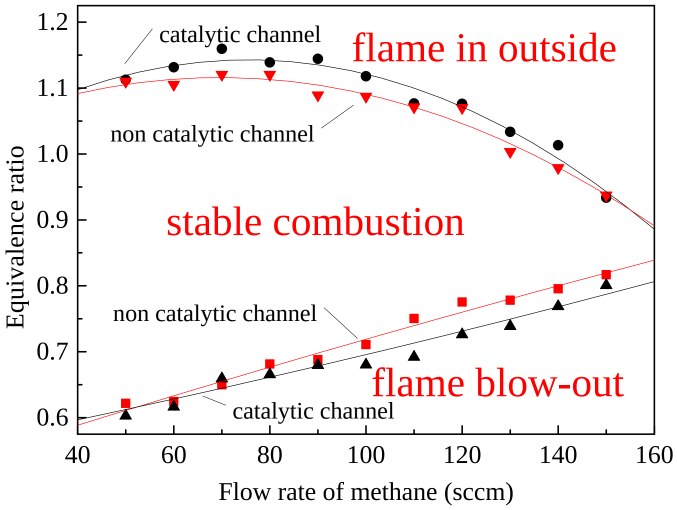

Fig. 3. Flammability limits of micro channel for different flow rates of methane.

Fig. 4 is the infrared photos of the outer wall at the equivalence ratio of 1 and inlet velocity of 0.5 m/s, Fig. 4 (a) and Fig. 4 (b) are catalytic and non-catalytic channels respectively. The bright region in the pictures means higher temperature, from which it can be seen that there is no obvious higher temperature, and the temperature distribution is uniform in catalytic channel. On the contrary, there is an obvious higher temperature in the non-catalytic micro channel, and its surface temperature distribution is not uniform, the higher temperature of which is concentrated in the channel upstream.

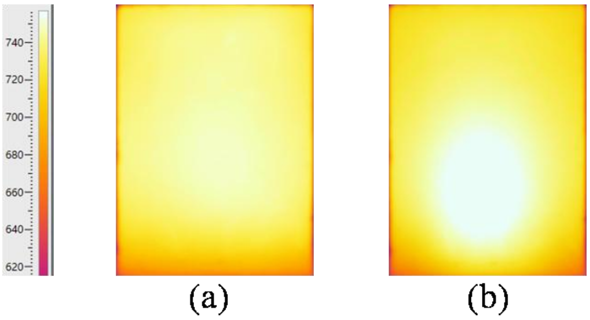

Fig. 4. Infrared images of the outer wall: （a）catalytic channel, （b）non-catalytic channel.

3.2. *Effect of equivalence ratio*

In order to study the effect of equivalence ratio on combustion characteristics of methane-air, inlet velocity is kept constant, and the centerline wall temperature profiles of outer wall in catalytic micro channel for different equivalence ratio from 0.6 to 1.1 at the inlet velocity of 0.5 m/s are obtained in Fig. 5. The small figure is the blowup of the circular region in Fig. 5. It can be seen that when the equivalence ratio is less than 0.9, the centerline temperature of the outer wall increase gradually with the increase of the equivalence ratio, and the highest temperature of outer wall is at the equivalence ratio of 0.9. The reasons lies in that methane is not enough in the fuel-lean condition, which made excess oxygen occupy space on the Pt and inhibiting the catalytic reaction on the inner surface, and the gas phase reaction is the main reaction. With the increase of equivalence ratio, the concentration of methane in the channel increases, which can release more heat during the combustion process, thus, makes the centerline temperature of the outer wall increase gradually. When the equivalence ratio is 1, the centerline temperature of the outer wall begins to decrease. The reason is that at this equivalence ratio, surface reaction has been strengthened, and heat released by the catalytic surface increase accordingly due to the existence of a catalyst, while fuel consumption and some other reasons lead to suppression of the gas phase reaction in the channel. The intensity of gas phase reaction within the channel weakened, and the temperature of the high temperature region decrease, resulting in the decrease of the centerline temperature of outer wall. When the equivalence ratio is 1.1, methane is in excess and insufficient air in the channel leads to the weakened gas phase reaction. This leaves less oxygen on the surface of the channel and makes surface reaction difficult to start. Besides, the excessive methane will take some heat away from the channel. Thus, temperature of the outer wall drops. In addition, the centerline temperature profiles have an intersection at the equivalence ratio of 1 and 1.1, and the point of intersection is 9 mm away from the channel entrance. This is because gas phase reaction and surface reaction occur simultaneously in the channel near the entrance at the equivalence ratio of 1, which makes the temperature rise and consumes more fuel during the reaction, thus leads to reduction of reaction intensity downstream of the channel. However, the reaction intensity near the entrance at equivalence ratio of 1.1 is low, making more fuel participate in reaction in the downstream of the channel, so the downstream centerline temperature at equivalence ratio of 1.1 is higher than the equivalence ratio of 1.

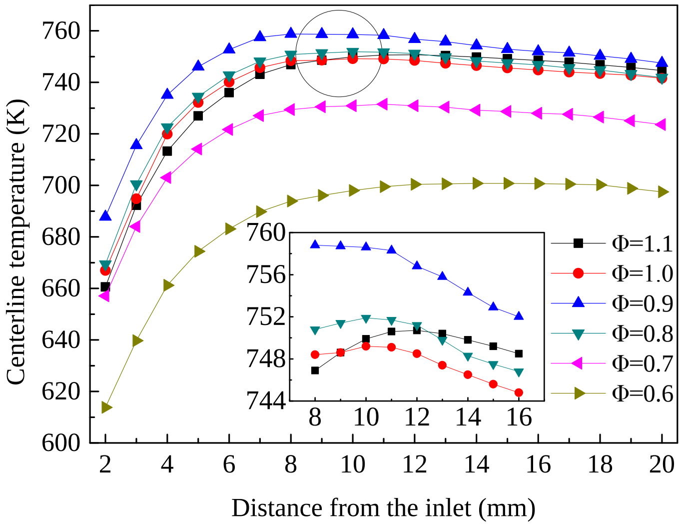

Fig. 5. Centerline temperature profiles of outer wall for different equivalence ratios along the catalytic micro channel.

Fig. 6 is the centerline temperature profiles of outer wall in the non-catalytic micro channel for different equivalence ratios under the same test condition. From Fig.6, with the increase of equivalence ratio, the changing tendency of temperature for non-catalytic micro channel is basically the same as that of the catalytic channel, but the temperature in the downstream drops quicker than that of catalytic micro channel. In addition, when the equivalence ratio is the same, the highest temperature of non catalyst micro channel is higher than that of the catalytic channel. For example, the highest temperature of catalyst micro channel is 758.8K which is lower than 789K of the non-catalytic micro channel at the equivalence ratio of 0.9. This is because there is no effect of surface reaction in the non-catalytic channel, and the intensity of gas phase reaction is the most violent, which leads to the most of heat release. But in the downstream of the channel, there is no heat release from surface reaction, which results in the quicker dropping of temperature compared to the catalytic channel. Therefore, adding catalyst in the micro channel can not only reduce the highest centerline temperature, but also gives a uniform temperature distribution on the outer wall.

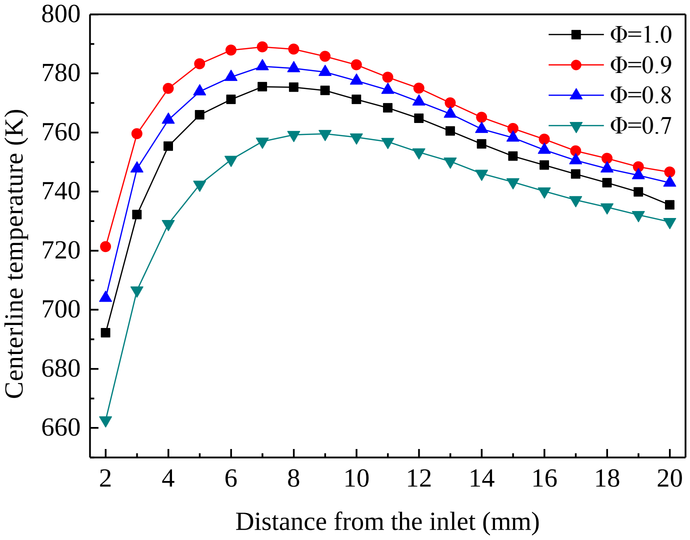

Fig. 6. Centerline temperature profiles of outer wall for different equivalence ratios along the non-catalytic micro channel.

3.3. Effect of inlet velocity

In order to study the influence of inlet velocity on the catalytic combustion characteristics of methane-air, the centerline temperature profiles of outer wall in catalytic micro channel for different inlet velocities from 0.4 m/s to 0.8 m/s at the equivalence ratio of 1 are obtained in Fig. 7, including catalytic side and non-catalytic side. It can be seen that the centerline temperature of the outer wall increases gradually with the increase of inlet velocity. This is because with the increase of inlet velocity, more methane participates in the reaction at the same time, and more heat will be released. In addition, the point of highest temperature moves towards the downstream of the channel gradually with the increase of inlet velocity, which indicates that, the position of the flame in the channel shifts towards the downstream of the channel gradually. In the downstream of the channel, the drop slope of the temperature profile in the catalytic side drops with the increase of the inlet velocity. The reason is that, all reaction components spread more quickly to the downstream with the increase of inlet velocity, leading to the surface reaction in the downstream of the channel been strengthened, and meanwhile releasing more heat, so that the slope of the centerline temperature decreases with the increase of inlet velocity. Compared with the catalytic side, the drop slope of the centerline temperature profiles in the non-catalytic side in the downstream of the channel is quicker than that of the catalytic side. This is because, in the downstream of the channel, there is no heat released from surface reaction on the inner surface in the non-catalytic side. But the drop slope of the temperature profiles in non-catalytic side is consistent. With the increase of the inlet velocity, the temperature difference between the adjacent velocity decreases gradually. The reason is that the residence time of methane in the channel is shortened as the inlet velocity increases, resulting in the degree of reaction and the release of heat are relatively weakened. Besides, the heat is blown out of the channel more quickly. As a result, the temperature differences between adjacent inlet velocities decreases.

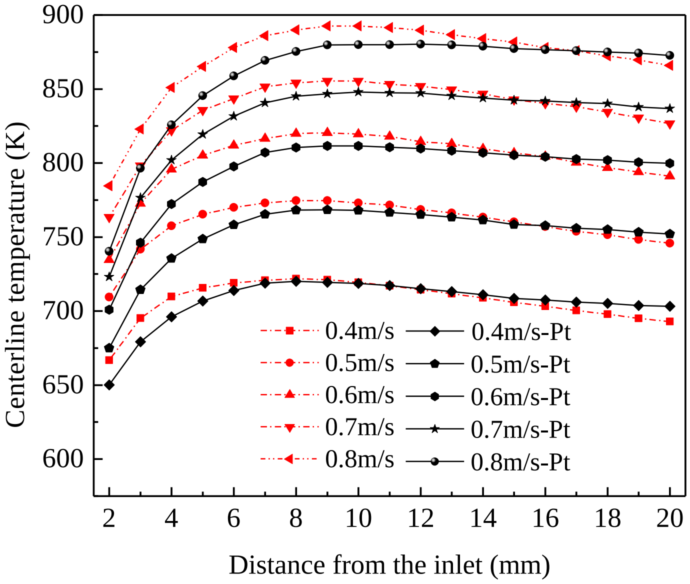

Fig. 7. Centerline temperature profiles of outer wall for different sides in catalytic channel.

Comparing the temperature profiles on both sides of the channel, it can be seen that the highest temperature of catalytic side is lower than the non-catalytic side at the same velocity, and with the variation of the distance from the inlet, the centerline temperature of catalytic side is lower than that of the non-catalytic side at first then higher than that of non-catalytic side. This is because, in the upstream of the channel, methane participates in surface reaction, which will consume some methane meanwhile inhibiting the gas phase reaction near the internal wall, which makes the temperature of catalytic side in the upstream lower than that of non-catalytic side. However, in the downstream of the channel, surface reaction and gas phase reaction are weakened together, and the heat release is also reduced gradually, which leads to the drop in temperature. In the catalytic side, there is still some remaining methane which has not reacted completely and would participate in the surface reaction in the downstream of the channel, which in effect releases some heat. But there is no surface reaction in the non-catalytic side, so the temperature in the catalytic side in the downstream of the channel is higher than that of non-catalytic side. Fig. 8 is the average temperature of the outer wall on both sides of the catalytic micro channel. It shows that with the increase of the inlet velocity, the average temperature of the outer wall increases gradually, but the rising slope decreases gradually, which indicates that the temperature difference between the adjacent inlet velocities decreases gradually, which is consistent with the above conclusion. The average temperature between different sides of the channel at the same inlet velocity is not big, which indicates that one catalytic surface in the channel has little impact on the average temperature distribution of the outer wall for different inlet velocities. The reason is that although the highest temperature of catalytic side is lower than that of non-catalytic side, because of the existence of the catalyst, the reaction area of catalytic side is larger than that of non-catalytic side. Therefore, the heat release for both sides at the same time is basically the same and the average temperature difference is not very big.

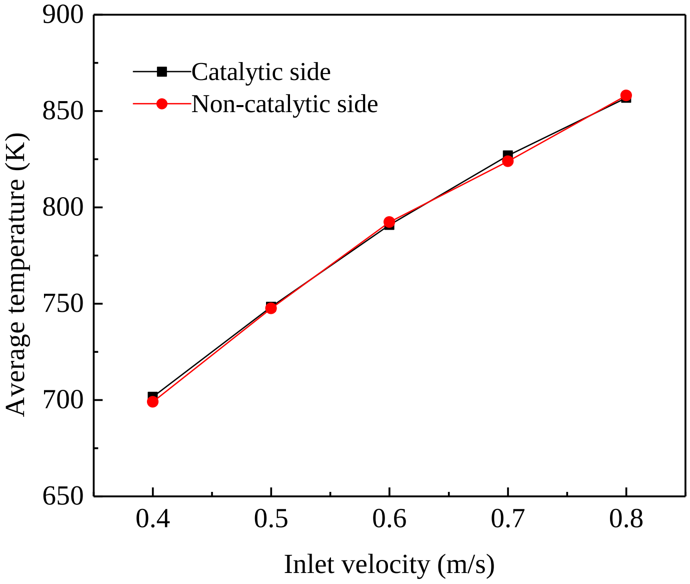

Fig. 8. Average temperature profiles of the outer wall for different inlet velocities.

To study the effect of the addition of catalyst on combustion characteristics for different inlet velocities, the experiment was carried out at the equivalence ratio of 1, and varying inlet velocity of premixed gas in the micro channel. Fig. 9 is the centerline temperature profiles of non-catalytic side in the catalytic channel and non-catalytic channel for different inlet velocities. Making a comparison between the centerline temperatures of the two types of micro channel, it can be seen that with the increase of inlet velocity, the centerline temperature of catalytic micro channel is higher than that of the non-catalytic micro channel at first, and then has an overlap, finally lower than non-catalytic micro channel. It is because that when the inlet velocity is low, the longer residence time of the fuel in the channel can provide sufficient reaction time on the catalyst surface, which will consume a lot of oxidizer at the same time, and suppress the gas phase reaction near the Pt, but its heat release can promote the gas phase reaction on the other side. When the inlet velocity is higher, the residence time of mixed gas in the channel is short, which will reduce the reaction time of methane in the channel accordingly, and methane will be blown out of the channel more easily. But the existence of the catalyst will absorb some methane on the surface, and its heat release will promote the gas phase reaction near the Pt. However, its consumption of the methane will inhibit the gas phase reaction on the other side of the wall, which leads to the centerline temperature of catalytic channel lower than that of the non-catalytic micro channel.

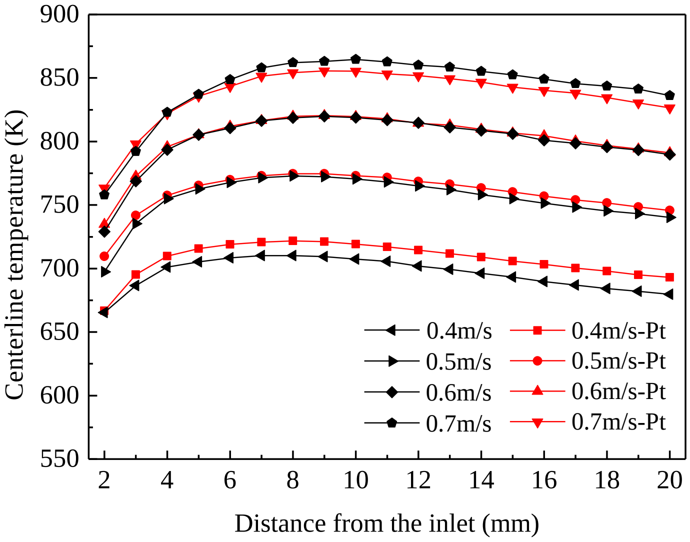

Fig. 9. Centerline temperature profiles of outer wall for catalytic and non-catalytic channel at different inlet velocities.

3.4. Analysis of exhaust gas

A flue gas analyzer was used to analyze the main components of the exhaust gas. The exhaust gas in catalytic and non-catalytic channel for different inlet velocities at the equivalence ratio of 1 was measured as shown in Fig. 10, which contains the volume fraction of CO, CO_2 and total C in the combustion products. The results showed that with the increase of inlet velocity, the volume fraction of CO_2 in combustion products decrease gradually and the volume fraction of CO decreases at first and then increase in the non-catalytic channel. When catalyst was added into the channel, the volume fraction of CO_2 in combustion products decreases gradually and the volume fraction of CO decreases at first and then increases in catalytic channel with the increase of the inlet velocity. Analyzing the total C in combustion products can explain the degree of combustion reaction indirectly. Making a comparison of total C in combustion products between the catalytic and non-catalytic channel, it can be seen that the volume fraction of total C in catalytic micro channel is significantly higher than that of non-catalytic micro channel, which indicates that the existence of the catalyst can improve the combustion degree for methane-air, that is to say, improves the combustion efficiency.

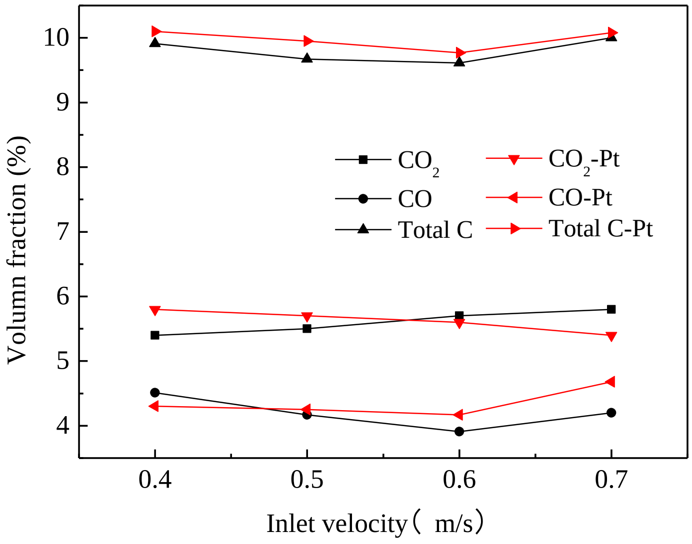

Fig. 10. The main components of the exhaust gas for different inlet velocities.

3.5. Effect of channel height

Channel height has an important impact on the combustion characteristics in the micro channel. In order to study the impact of channel height on the catalytic combustion characteristics, the same material and processing methods were used to process the catalytic micro channel with the height of 2mm and 2.5mm respectively. The centerline temperature profiles of the outer wall for different inlet velocities and different channel heights at the equivalence ratio of 1 are seen in Fig. 11. It can be seen that when the height of the channel is the same, the temperature difference between the temperature near the entrance and the highest temperature increases gradually with the increase of the inlet velocity. The main reason is that before the temperature reaches the highest, gas phase reaction and surface reaction are strengthened together, and when it reaches the highest temperature, the reactions and the heat release are the most intense. However, in the downstream of the channel, the intensity of the reaction decreases, resulting in a corresponding decrease in the outer wall temperature. With the increase of inlet velocity, more mixed gas need to absorb heat to participate in the reaction near the entrance of the channel, but because of the reaction region of the high temperature away from the entrance of the channel, the distance traveled by heat to the upstream increases, which results in a corresponding increase in thermal resistance. Therefore, the temperature difference increases. When the inlet velocity is constant, the centerline temperature of outer wall increases with the increase of the channel height. The reason is that the energy been imported to the channel increase with the increase of channel height at the same time, and more methane can participate in the reaction, which leads to a corresponding increase in heat release, so the centerline temperature of the outer wall increases gradually. The temperature profiles in the downstream of the channel are similar at different channel heights and inlet velocities, and the temperature drop slopes are basically the same.

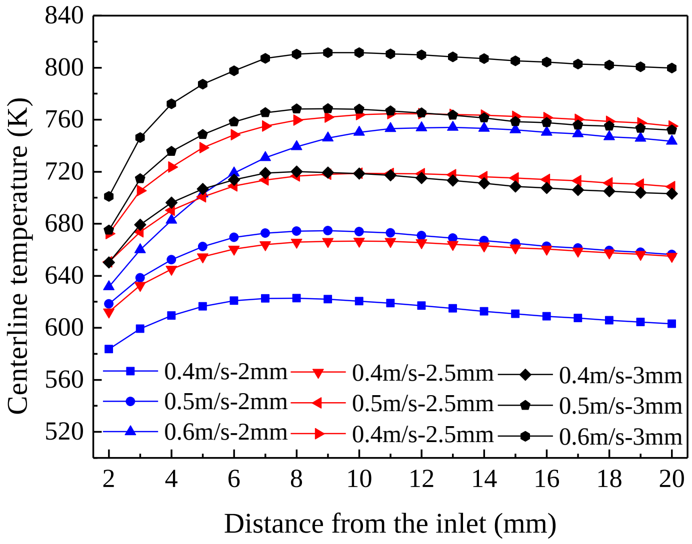

Fig. 11. Centerline temperature profiles of outer wall for different channel heights.

4. Conclusions

In this paper, experimental study on the catalytic combustion characteristics for methane-air in a rectangular micro channel was carried out, and some important conclusions were obtained as follows:

(1) With the increase of the flow rate of methane, the flammability limits under fuel rich conditions increase at first and then decrease and the flammability limits under fuel lean conditions decrease gradually in both catalytic and non-catalytic micro channels. The flammability limits are obviously improved when a catalyst is added into the channel.

(2) When the height of the micro channel is 3mm, no matter there is catalyst in the micro channel or not, the centerline temperature of outer wall increases at first and then decreases with the increase of equivalence ratio. And the maximum heat release and the highest centerline temperature of the outer wall are obtained at the equivalence ratio of 0.9.

(3) The centerline temperature of outer wall increases with the increase of inlet velocity when the equivalence ratio is the same, and the point of highest temperature shifts to the downstream of the channel gradually.

(4) Combustion intensity in the channel increases with the increase of channel height at the same inlet velocity. The addition of catalyst in the channel can also improve methane conversion.

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Figure captions

Fig. 1. Schematic diagram of experimental system.

Fig. 2. Section structure of catalytic micro channel.

Fig. 3. Flammability limits of micro channel for different methane flow rates.

Fig. 4. Infrared images of the outer wall: （a）catalytic channel, （b）non-catalytic channel.

Fig. 5. Centerline temperature profiles of outer wall for different equivalence ratios along the catalytic micro channel.

Fig. 6. Centerline temperature profiles of outer wall for different equivalence ratio along the non-catalytic micro channel.

Fig. 7. Centerline temperature profiles of outer wall for different sides in catalytic channel.

Fig. 8. Average temperature profiles of the outer wall for different inlet velocities.

Fig. 9. Centerline temperature profiles of outer wall for catalytic and non-catalytic channel at different inlet velocities.

Fig. 10. The main components of the exhaust gas for different inlet velocities.

Fig. 11. Centerline temperature profiles of outer wall for different channel heights.