

Investigation on the influencing parameters of methane catalytic ignition temperature in a Micro –Channel

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ABSTRACT

By using detailed reaction mechanism of CH₄ catalytic oxidation on Pt surface, the catalytic oxidation of CH₄-Air mixtures was investigated numerically in a micro-channel, focusing on the effects of operating conditions on catalytic ignition of methane. The results show that with the increase of the equivalence ratio, the opportunity of CH₄ getting adsorbed by the active sites increases, resulting in a lower ignition temperature of methane. The residence time of CH₄ in the channel is negatively correlated to the inlet velocity, and a lower inlet velocity is good for CH₄ ignition. Increase of surface catalyst loading density provides more active sites for CH₄ adsorption and reaction, thus lowers the ignition temperature of methane. When keeps the flow rate of CH₄-Air mixture as constant, oxygen can hardly desorb from the active sites at high pressure condition, which inhibits the adsorption of CH₄, and leads to a high ignition of methane temperature.

1. INTRODUCTION

In recent years, with the development of micro fabrication technology and micro-electro-mechanical systems (MEMS), more and more researchers are aware of micro burners as a promising type of portable energy device. Micro burners have many advantages, such as the simple structure, long working life, easy fuel replacement, and providing powers from a number of microwatts to hundreds of watts. They have been widely applied to the military, aerospace, chemical analysis, biomedical, environmental monitoring, digital electronic products and other fields. The combustion stability and efficiency of micro burners directly affect the performance of micro-electromechanical systems. It is necessary to carry out an in-depth study of the combustion characteristic of micro-burner. Compared to conventional burners, the dimensions of micro-burner's are quite small, leading to a quenching distance. In the millimeter scale channels, it is difficult to maintain a stable flame, although it has been proved that in specially treated

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channels with widths of 0.5 and 0.75 mm, gas phase combustion of methane can occur. Due to the small micro-burner size and large surface area, heat loss is generally huge. When the space size is smaller than 1 mm, the gas phase reaction can be ignored, because of the serious heat and radical losses. The micro channel size has a significant effect on ignition and combustion of the internal gas. How to achieve rapid ignition and stable combustion in micro-scale conditions has become a key problem of the MEMS combustion technology.

Recently, fuel combustion stability and combustion efficiency affecting parameters make up of the main research of methane/air catalytic combustion in micro channel. Lee et al established a simplified theoretical model of the micro burner and analyzed the heat loss and flame stability of millimeter combustion chamber. Through numerical simulation about the different diameters of the combustion chamber on the combustion of methane / air, Hua et al studied the effects of different wall heat transfer conditions and wall thermal conductivity on micro-combustion. Zhong Beijing et al adopted the CFD and DETCHEM in combination to study methane catalytic combustion in a heat exchanger burner, their results show that the catalytic combustion of methane can be helpful to stable and highly efficient transition. John Mantzaras et al adopted detailed elementary reaction mechanism to investigate methane catalytic combustion over Pt numerically. Jiao Jian et al studied the catalytic combustion of premixed methane/air in a micro channel using computational fluid dynamics software Fluent and chemical kinetics software DETCHEM, and the results show that the inlet flow rate has a great impact on the catalytic reaction. The above scholars mainly studied methane / air catalytic combustion stability and combustion efficiency, while the study on characteristics of methane catalytic ignition temperature is still relatively rare. At present, achieving catalytic ignition of methane mainly depends on two ways: 1) The burner wall temperature lights the fuel mixture gas; 2) Directly preheating the inlet of methane / air. In this paper, we use numerical methods to study the ignition conditions of fuel-lean mixture of CH₄/air, focusing on the effects of operating conditions on catalytic ignition temperature of CH₄.

2 PHYSICAL AND MATHEMATICAL DESCRIPTIONS

2.1 Physical model

The configuration of the 3-D micro-tube adopted in this paper is shown in Fig.1. The internal radius of the tube r_0 is 0.5 mm, the thickness of the wall b is 0.1 mm and the whole length of the tube L is 10 mm. Pt catalysts is assumed to be uniformly deposited on the smooth inner-wall with a density of $2.72 \times 10^{-8} \text{ kmol/m}^2$. The center of the tube entrance locates at the origin of coordinates, and the inlet velocity of the mixture gas is uniform at the entrance of the tube and positive with x-axis. Catalytic combustion occurs at the surface of catalyst, which the gas reaction rate is quite slow compared to gaseous combustion. It can be difficult to ignite by sparks. Continuous-heating is necessary for successful ignition. Therefore, the inlet temperature is simply fixed at 790K, which is approximate to the ignition temperature of the present mixture with equivalence ratio 0.2 (CH₄: 2vol. %, O₂: 20vol. %, N₂:76vol. %). The Reynolds number based on the inlet properties (inlet velocity is 1 m/s) is about 25, leading to a laminar flow in the micro-tube. The coupled heat transfer happens between the solid and the fluid phase.

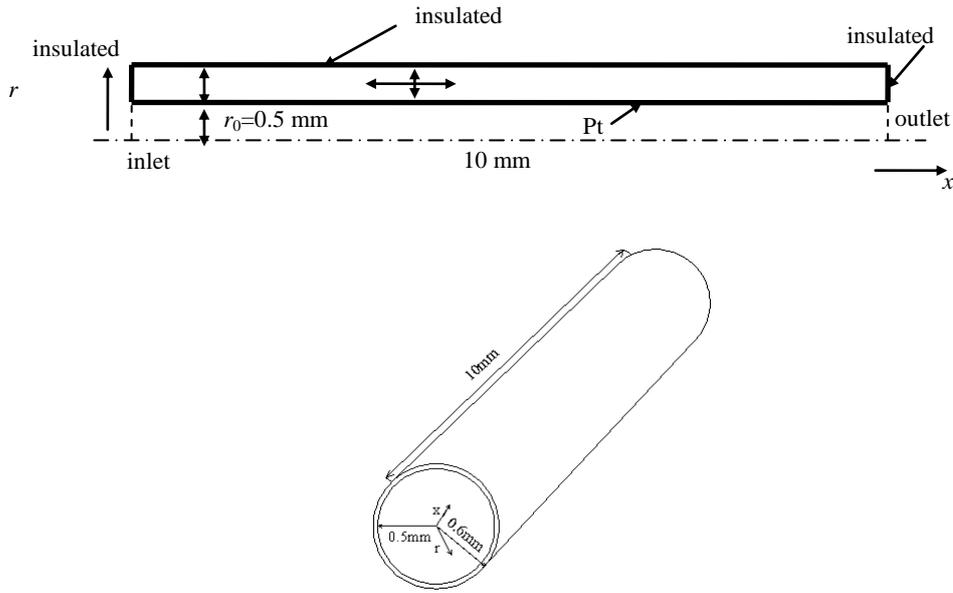


Fig.1 Schematic of the micro-tube configuration

2.2 Numerical models

Though the micro-tube is millimeter-scale, its characteristic size is still much bigger than the average free distance of the reactant molecules. So it is reasonable to adopt continuum model in this paper. In addition, the body force term, dissipations and radiation of the gaseous species are neglected. The main governing equations are as follows.

Continuum equation,

$$\frac{\partial(\rho u)}{\partial x} + \frac{1}{r} \frac{\partial(r \rho v)}{\partial r} = 0 \quad (1)$$

Momentum equation,

$$\frac{\partial(\rho u u)}{\partial x} + \frac{\partial(\rho u v)}{\partial r} = -\frac{\partial P}{\partial x} + \frac{\partial}{\partial x} \left[\frac{4}{3} \mu \frac{\partial u}{\partial x} - \frac{2}{3} \mu \frac{1}{r} \frac{\partial(r v)}{\partial r} \right] + \frac{1}{r} \frac{\partial}{\partial r} \left[\mu r \left(\frac{\partial v}{\partial x} + \frac{\partial u}{\partial r} \right) \right] \quad (2)$$

$$\begin{aligned} \frac{\partial(\rho u v)}{\partial x} + \frac{\partial(\rho v v)}{\partial r} = & -\frac{\partial P}{\partial r} + \frac{\partial}{\partial x} \left[\mu \left(\frac{\partial v}{\partial x} + \frac{\partial u}{\partial r} \right) \right] \\ & + \frac{\partial}{\partial r} \left[\frac{4}{3} \mu \frac{\partial v}{\partial r} - \frac{2}{3} \mu \left(\frac{\partial u}{\partial r} + \frac{v}{r} \right) \right] + \frac{2\mu}{r} \left(\frac{\partial v}{\partial r} - \frac{v}{r} \right) \end{aligned} \quad (3)$$

Energy equation of the gas phase,

$$\begin{aligned} \frac{\partial(\rho u h)}{\partial x} + \frac{\partial(\rho v h)}{\partial r} = & \frac{\partial}{\partial x} \left(\lambda \frac{\partial T}{\partial x} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left(r \lambda \frac{\partial T}{\partial r} \right) \\ & + \frac{\partial}{\partial x} \left(\sum_{k=1}^{K_i} h_k \rho D_k \frac{\partial Y_k}{\partial x} \right) + \frac{\partial}{\partial r} \left(\sum_{k=1}^{K_i} h_k \rho D_k \frac{\partial Y_k}{\partial r} \right) \end{aligned} \quad (4)$$

The mixture-average diffusion coefficient of gaseous species k , D_k , is determined by

$$D_k = \frac{1}{\sum_{l=1, l \neq k}^{K_i} \frac{Y_l \bar{M}}{M_l D_{kl}}} \quad (5)$$

Species equation of the gas phase,

$$\frac{\partial(\rho u Y_k)}{\partial x} + \frac{\partial(\rho v Y_k)}{\partial r} = \frac{\partial}{\partial x}(\rho D_k \frac{\partial Y_k}{\partial x}) + \frac{1}{r} \frac{\partial}{\partial r}(r \rho D_k \frac{\partial Y_k}{\partial r}) \quad (k=1, 2, \dots, K_i) \quad (6)$$

Energy equation of the solid wall,

$$\frac{\partial}{\partial x}(\lambda_s \frac{\partial T}{\partial x}) + \frac{1}{r} \frac{\partial}{\partial r}(r \lambda_s \frac{\partial T}{\partial r}) = 0 \quad (7)$$

Finally, the ideal gas and caloric equation of state are,

$$p = \rho RT \sum_{k=1}^{K_i} \frac{Y_k}{M_k} \quad (8)$$

$$h = h_0(T_0) + \int_{T_0}^T c_p dT \quad (9)$$

D Diffusion coefficient (m²/s), p pressure, u, v Axial, radial velocities (m/s), h Total enthalpy of the gas phase (J/K_g), K_i, K_j Total number of gaseous species and surface species, r Radial coordinate (m), x Axial coordinate (m), ρ Density (Kg/m³), Thermal conductivity(W/m/K), Y Mass fraction, μ Viscosity (Pa·s), Φ Methane/oxygen equivalence ratio.

2.3 Reaction mechanism

In a millimeter-scale catalytic combustor, homogeneous reaction is generally ignored, since it will be inhibited by the use of catalysts. So only a heterogeneous mechanism is included in the calculation of this paper. The detailed elementary heterogeneous mechanism of methane over Pt established by Deutschmann et al. is adopted here to predict the combustion process. Nine adsorption reactions, nine surface elementary reactions and six desorption reactions are included in this mechanism (see in table 1), ten gas phase species (CH₄, H₂, O₂, CO₂, CO, H₂O, H, O, OH, N₂) and eleven surface species (Pt(s), H(s), H₂O(s), OH(s), CO(s), CO₂(s), CH₃(s), CH₂(s), CH(s), C(s), O(s)) are used to describe the detailed process during the combustion of methane. The mechanism is commonly used in CFD studies of methane catalytic combustion. Its applicability and reliability had been proved by literature.

Table 2.1 24-step-elementary reaction mechanism of CH₄ catalytic combustion

Reaction	A, S ₀	β	E (KJ/mol)	ε _i , μ _i ^a
1 H ₂ +2Pt(s)=>2H(s)	4.60×10 ⁻²			μ _{Pt(s)} =-1

2	$2\text{H(s)} \Rightarrow 2\text{Pt(s)} + \text{H}_2$	3.70×10^{21}	0.0	67.4	$\varepsilon_{\text{H(s)}}=6$
3	$\text{H} + \text{Pt(s)} \Rightarrow \text{H(s)}$	1.00			
4	$\text{O}_2 + 2\text{Pt(s)} \Rightarrow 2\text{O(s)}$	1.81×10^{21}	-0.5	0.0	
5	$\text{O}_2 + 2\text{Pt(s)} \Rightarrow 2\text{O(s)}$	2.30×10^{-2}			
6	$2\text{O(s)} \Rightarrow \text{O}_2 + 2\text{Pt(s)}$	3.70×10^{21}	0.0	213.2	$\varepsilon_{\text{O(s)}}=60$
7	$\text{O} + \text{Pt(s)} \Rightarrow \text{O(s)}$	1.00			
8	$\text{H}_2\text{O} + \text{Pt(s)} \Rightarrow \text{H}_2\text{O(s)}$	0.75			
9	$\text{H}_2\text{O(s)} \Rightarrow \text{H}_2\text{O} + \text{Pt(s)}$	1.00×10^{13}	0.0	40.3	
10	$\text{OH} + \text{Pt(s)} \Rightarrow \text{OH(s)}$	1.00			
11	$\text{OH(s)} \Rightarrow \text{OH} + \text{Pt(s)}$	1.00×10^{13}	0.0	192.8	
12	$\text{O(s)} + \text{H(s)} \Leftrightarrow \text{OH(s)} + \text{Pt(s)}$	3.70×10^{21}	0.0	11.5	
13	$\text{H(s)} + \text{OH(s)} \Leftrightarrow \text{H}_2\text{O(s)} + \text{Pt(s)}$	3.70×10^{21}	0.0	17.4	
14	$\text{OH(s)} + \text{OH(s)} \Leftrightarrow \text{H}_2\text{O(s)} + \text{O(s)}$	3.70×10^{21}	0.0	48.2	
15	$\text{CO} + \text{Pt(s)} \Rightarrow \text{CO(s)}$	8.40×10^{-1}			$\mu_{\text{Pt(s)}}=1$
16	$\text{CO(s)} \Rightarrow \text{CO} + \text{Pt(s)}$	1.00×10^{13}	0.0	125.5	
17	$\text{CO}_2\text{(s)} \Rightarrow \text{CO}_2 + \text{Pt(s)}$	1.00×10^{13}	0.0	20.5	
18	$\text{CO(s)} + \text{O(s)} \Rightarrow \text{CO}_2\text{(s)} + \text{Pt(s)}$	3.70×10^{21}	0.0	105.0	
19	$\text{CH}_4 + 2\text{Pt(s)} \Rightarrow \text{CH}_3\text{(s)} + \text{H(s)}$	1.00×10^{-2}			$\mu_{\text{Pt(s)}}=0.3$
20	$\text{CH}_3\text{(s)} + \text{Pt(s)} \Rightarrow \text{CH}_2\text{(s)} + \text{H(s)}$	3.70×10^{21}	0.0	20.0	
21	$\text{CH}_2\text{(s)} + \text{Pt(s)} \Rightarrow \text{CH(s)} + \text{H(s)}$	3.70×10^{21}	0.0	20.0	
22	$\text{CH(s)} + \text{Pt(s)} \Rightarrow \text{C(s)} + \text{H(s)}$	3.70×10^{21}	0.0	20.0	
23	$\text{C(s)} + \text{O(s)} \Rightarrow \text{CO(s)} + \text{Pt(s)}$	3.70×10^{21}	0.0	62.8	
24	$\text{CO(s)} + \text{Pt(s)} \Rightarrow \text{C(s)} + \text{O(s)}$	1.00×10^{18}	0.0	184.0	

2.4 Calculation method and model validation

According to the structure of the micro-cavity, the hexahedral form of structured grid is adopted whose grid size is taken as 0.1mm. Total number of grid independence check is done by the calculation results. The solid wall and the fluid region have 76250 grid cells. To continue to increase the grid to 86240, a calculation result has the deviation of 0.8%. Because $\text{Re} < 2300$, Laminar flow model and component transmission model is employed. The method for solving the problem is the SIMPLE algorithm. Import speed inlet boundary (the residence time of the gas mixture in the tube is much larger than the reaction time) and pressure outlet boundary are put to use. Inlet and outlet pressure are an atmospheric pressure. Outer wall is insulated and inner wall uses the coupling wall. No rust steel makes up of burner material. Gas diffusion puts mixed the average diffusion model in use and does not consider the thermal diffusion.

Methane ignition temperature was measured in the experiment which had been done by Behrendt et al and the numerical model predictions of this paper were compared. During the Experiment, a certain equivalence ratio mixed gas of methane and oxygen by 94 Vol. % nitrogen diluted flows to platinum plate with the speed of 8 cm/s vertically. This flow forms a stagnation point flow. The platinum plate connected to the current generates heat and the thermocouple (two platinum wires) is used to monitor the temperature. If the temperature suddenly jumps, methane ignition will occur. The temperature which jumps in front of it is the ignition temperature.

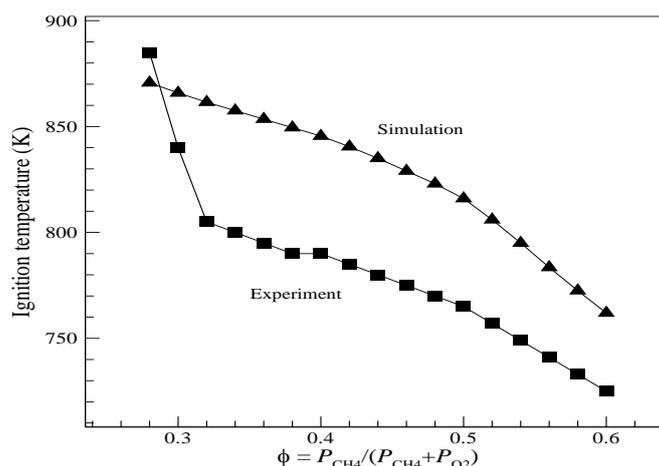


Fig.2 The numerical prediction ignition temperature comparison of experimental results

Definition of methane / air equivalence ratio:

$$\phi = \frac{P_{CH_4}}{P_{CH_4} + P_{O_2}} \quad (10)$$

In the numerical simulation, the outer wall is set to adiabatic conditions. The mixed gas component concentration of the inlet is consistent with the experiment. The inlet gas temperature is gradually increased until the methane happens with catalytic ignition. At the same time, the methane inlet temperature is ignition temperature. The qualitative trend of Methane ignition temperature with increasing concentrations is same and the largest gap of it is approximately 4.5% from quantity, so catalytic combustion reaction kinetics model used is correct in the paper.

3 RESULTS AND DISCUSSION

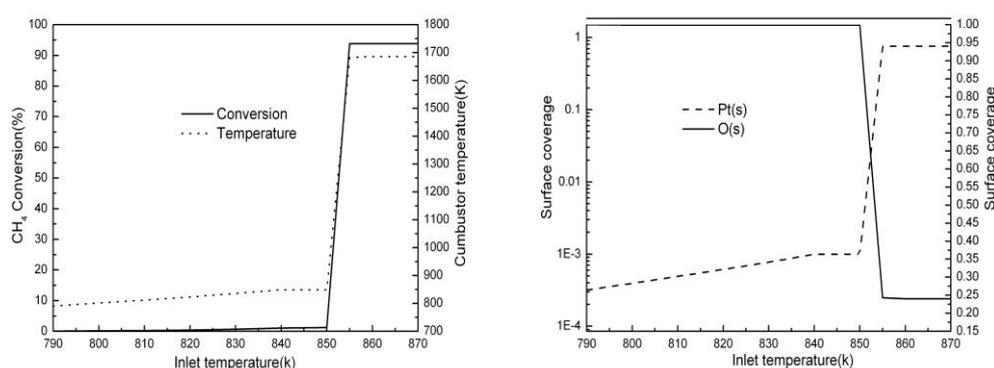
3.1 Calculation

CH₄, O₂ and N₂ constitute the mixed gas. N₂ is an inert gas and does not participate in the reaction. The reaction O₂ + 2Pt (s) → 2O(s) and 2O(s) → O₂ + Pt(s) has the most significant impact on CH₄ combustion through sensitivity analysis. To eliminate the impact of O₂, the O₂ is excess and the volume fraction is constant at 20%. Reaction conditions are to study the impact of methane catalytic ignition temperature characteristics by changing other conditions.

3.2.1 CH₄ catalytic ignition in micro circular tube

From Fig.3 we can see that after the mixed gas was preheated to 790K

(equivalence ratio 0.6, the wall surface of the catalyst load density $2.72 \times 10^{-8} \text{ kmol/m}^2$, mixed gas pressure 1atm) into the burner, gas molecules flow in the axial direction and spread to the catalytic wall in radial condition. Compared to CH_4 , O_2 has a larger volume fraction. Pt catalyst have a higher selectivity of the O_2 molecule than CH_4 molecule, so the wall surface of active ingredient (Pt (s)) is almost wholly covered by O(s), only a small amount of CH_4 was adsorbed and oxidated. With the increase of inlet gas temperature, high-temperature gas passes the heat to the catalytic wall surface, so the wall temperature will be increased. The high wall temperature strengthens the O_2 desorption, so O(s) decreases in the coverage of wall and increases the amount of vacancy active center. When the inlet gas temperature is increased to 855K, CH_4 ignition occurs and the temperature inside the reactor swells to 1685K. At the same time, coverage of the wall surface of the active ingredient is also changed greatly.

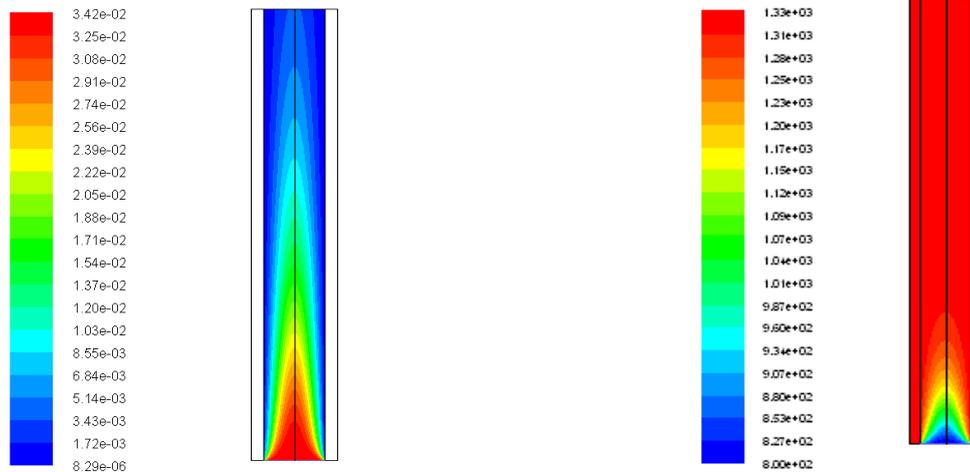


(a) Methane conversion and combustor Temperature with different inlet temperature

(b) Surface coverage with different inlet temperature

Fig.3 Methane catalytic combustion characteristics with inlet temperature

Catalytic reactions require two prerequisites: ① Temperature reaches above the activation temperature; ② A certain amount of active centers. As can be seen from the above calculation results, The micro scale CH_4 catalytic ignition is hard to occur, because the noble metal catalyst Pt have the high adsorption of O_2 than CH_4 , resulting vacancy active center covered by O(s) mostly. CH_4 adsorption and reaction is suppressed, therefore CH_4 ignition requires a higher temperature. The condition is shown in the above: CH_4 ignition temperature is 855K. At this time, the methane conversion rate and the chamber temperature increase dramatically. O(s) covering wall surface is a sharp decline to participate in the reaction and the corresponding Pt (s) rises which is conducive to the catalytic ignition of methane.



(a) Mass fraction of methane distribution of combustor (b) Temperature distribution of combustor

Fig.4 Methane catalytic combustion characteristics after ignition

As you can see from Fig.4, the burner temperature distribution is more obvious in the importing location owing to the thermal conductivity. Residence time of methane in the burner is sufficient to maintain the methane catalytic combustion. Methane catalytic oxidation reaction happens in front of the burner completely, high temperature is conducive to oxygen desorption from the wall activity center which is conducive to the catalytic oxidation of methane.

3.2.2 The effects of equivalence ratio on methane catalytic ignition

From Fig .5, the methane gas (inlet velocity 1m / s, pressure 1atm, the wall catalyst loading density $2.72 \times 10^{-8} \text{ kmol/m}^2$) is sent to reactor. Oxygen volume fraction is guaranteed to 20%, Average temperature of the burner is greatly changed owing to the different methane / oxygen equivalence ratio. Observing from the following figure, the combustion temperature is not much changing at the inlet temperature of 820K when the methane is not burnt. When the preheating temperature of the mixed gas increases, the gas whose equivalent ratio is 1 began to ignite. At this time, average temperature of the burner has a sharp rise and the methane concentration has a sharp decrease. At the equivalent ratio of 1, because the methane concentration is relatively large, the speed of the catalytic oxidation of methane accelerates. The reaction heat emit is greater than the heat dissipate, so the methane is ignited.

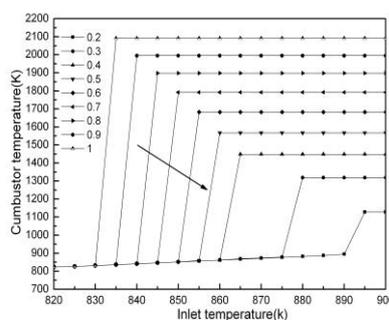


Fig.5 combustion temperature with different inlet temperature

As is shown in Fig.6 and Fig.7, with different equivalence ratio, the ignition temperature of methane is different. With the equivalence ratio of 0.2,0.3,0.4,0.5,0.6,0.7, 0.8,0.9,1, the ignition temperature of methane are 895k, 880k, 869k, 861k, 855k, 849k, 845k, 840k , 836k,respectively. Another curve means that the methane before ignition emits heat, the average temperature of the burner rise to some extent. Because the partial oxidation methane was occurred before ignition, combustion temperature is elevated, but the preheating temperature is less than the ignition temperature of methane.

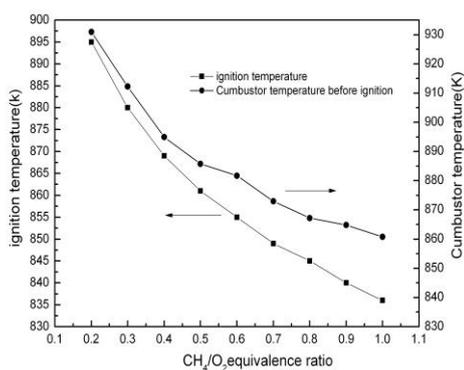


Fig.6 Ignition temperature with different equivalence ratio

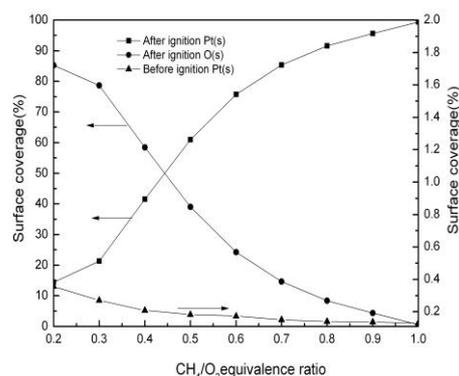


Fig.7 Surface coverage with different equivalence ratio

Methane ignition temperature decreases with increasing equivalence ratio. Methane/oxygen of large equivalent ratio represent big concentration of methane in the process of chemical reaction kinetics. The concentration of methane increases, this makes the chemical reaction speed accelerates and heat accumulation happens quickly. Under this condition, it makes the methane be on fire.

Displayed in graph, wall active ingredient coverage makes a big difference with equivalence ratio before ignition. Pt(s) component covering the wall only can be found to 0.1%-0.3%. The vast majority of active space center is occupied by O(s). Adsorption of methane is hindered and methane can not fully participate in the reaction which can not continue to emit heat. When the conditions are reached the ignition temperature in each equivalence ratio, the wall surface of Pt(s) component sharply increased, while the wall surface of O(s) quickly decreases. The elementary reactions $CH_4 + 2Pt(s) \Rightarrow CH_3(s) + H(s)$ play a major role in methane combustion. Methane has a sharp decline in the outlet surface and the methane conversion is close to 100%. Found from the figure, as the equivalence ratio increases, the active center on the wall surface is occupied by O(s) component more and Pt(s) component less. With the methane / air equivalence ratio gradually increases, the combustion burner temperature was gradually elevated. The wall surface of the primitive reaction $O_2 + 2Pt(s) \Rightarrow 2O(s)$ displays important. When the ratio is small, methane concentration is relatively more and oxygen concentration is less, so the wall activity center is occupied by O(s) more than Pt(s) component.

3.2.2 The effects of gas flow rate on methane catalytic ignition

The methane mixed gas (methane / air equivalence ratio 0.6, pressure 1atm, the wall catalyst loading density $2.72 \times 10^{-8} \text{ kmol/m}^2$) is sent to reactor. Oxygen volume fraction

is guaranteed to 20%, Average temperature of the burner is greatly changed owing to the different inlet velocity. Mixed gas flow rate are respectively 0.1m/s ,0.25m/s, 0.5m/s, 0.75m/s, 1m/s, 1.25m/s, 1.5m/ s, 2m/ s, and the corresponding ignition temperature of methane are 839k, 841k, 846k, 851k, 855k, 860k, 862k, 869k. From Fig.8, it can be found that a portion of the methane have oxidation reaction before the catalytic ignition of methane. When the preheating temperature of the methane reaches the ignition temperature, the sharp rise has burner's average temperature and the methane conversion rate is increased dramatically. It can be found from the figure that methane requires a high ignition temperature because of high inlet velocity.

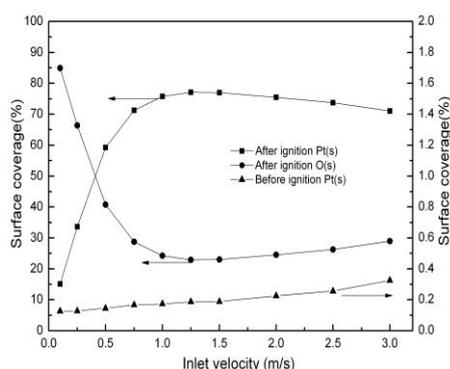


Fig.8 Ignition temperature with different inlet velocity

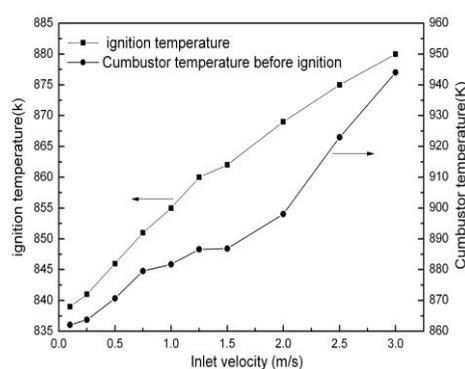


Fig.9 Surface coverage with different inlet velocity

With increasing the gas velocity, the residence time in the reactor is reduced. Because the reaction degree of methane decreases, the temperature inside the reactor reduces which require a high methane ignition temperature.

From Fig.9, active component covering the wall changes with the gas mixture velocity. Before ignition, the wall Pt(s) components occupy the active center which accounts for only 0.12%-0.23%, and the rest just are the O(s) component. When methane is ignited, Pt(s) on the wall surface of the active center component has a sharp increase and O(s) dropped quickly. As the flow rate increases, the primitive reaction $2O(s) \Rightarrow O_2 + Pt(s)$ has a big influence on it. The oxygen in the burner will reduce the residence time by increasing inlet velocity and the reaction will be strengthened to the Pt(s) direction. Thus the active center of Pt(s) component is gradually increased when the inlet gas flow rate continues to increase to the catalyst surface.

3.2.3 The effects of catalyst loading density on methane catalytic ignition

From Fig.10, The mixed methane / air (equivalence ratio 0.6, inlet pressure 1atm, inlet velocity 1m/s) are sent to reactor which Oxygen volume fraction is guaranteed to 20%. Ignition temperature of methane changes with the wall surface of the catalyst density. Wall catalyst Pt coverage density attains $2.72 \times 10^{-8} \text{ kmol/m}^2 \cdot \text{a}$. which is respectively 1, 2, 3, 4, 5, 6, 7, 8, 9, 10. Methane catalytic ignition temperature is respectively 855k, 816k, 795k, 780k, 770k, 761k, 754k, 748k, 743k, and 738k. As can be seen from Fig.10, with catalyst loading density increasing, the area that mixed gas contact catalyst surface per unit time will increase. This is helpful to catalytic oxidation of methane to emit heat, at the same time; it increases the adsorption time. With the starting wall surface of

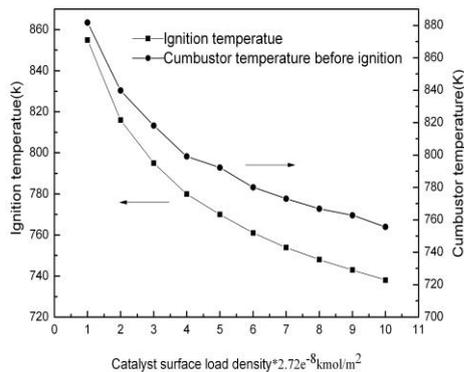


Fig.10 Ignition temperature with different catalyst loading density
Catalyst loading density increases, the surface Pt(s) increase correspondingly. This is conducive to primitive reactions $\text{O}_2 + 2\text{Pt}(s) \Rightarrow 2\text{O}(s)$ occur. In lower temperature, methane catalytic ignition can be easily happen.

3.2.3 The effects of pressure on methane catalytic ignition

From Fig.11, The mixed methane / air (equivalence ratio 0.6, inlet velocity 1m/s, the wall catalyst loading density $2.72 \times 10^{-8} \text{ kmol/m}^2$) are sent to reactor which Oxygen volume fraction is guaranteed to 20%. Ignition temperature of methane changes with the inlet pressure. Inlet pressure (atm) is respectively 1, 1.5, 2, 2.5, 3.5, 4, 4.5, and 5. Methane catalytic ignition temperature attains 855K, 862k, 869k, 874k, 878K, 882k, 886k, 889k, and 891k. As shown, when the preheating temperature does not reach the ignition temperature of methane, there is a small amount of methane which happens to catalytic oxidation. This just makes the reactor's temperature increase. At the same time, with the inlet gas pressure increases, methane catalytic ignition needs a high temperature.

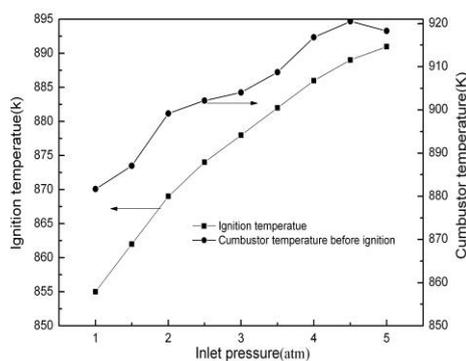


Fig.11 Ignition temperature with different inlet pressure

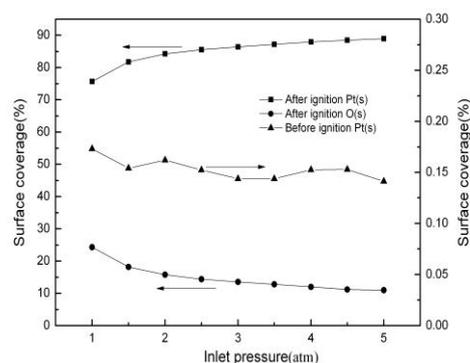


Fig.12 Surface coverage with different inlet pressure

When the inlet pressure increases at the constant inlet velocity, it is difficult for oxygen to have desorption from active center and diffuse gaseous phase. The vacancy active center decreases which lead to a high methane catalytic ignition temperature.

From Fig.12, the wall activity center Pt(s) account for only 0.15%-0.18% before ignition. After Methane catalytic ignition occurs, the wall surface of the active center Pt(s)

component has a sharp increase and O(s) component has a sharp drop. As the intake pressure increases, the wall surface of the active center Pt(s) initially rises quickly, and then tends to a constant value.

At the constant mass flow rate as $PV = \text{const}$, the results show that the ignition temperature basically does not change much with intake pressure at the same mass flow. When the quantity of gas is kept constant into the reactor per unit time, an increase in intake pressure represents a drop in inlet velocity. The intake velocity which is amplified is conducive to the ignition of methane, but the increase in pressure is not conducive to ignition of methane, the two effects offset each other.

4 CONCLUSIONS

(1) In the Micro –Channel, the oxygen adsorption capacity by the active centers of the wall surface is stronger than methane. This just results in a higher ignition temperature in low concentrations of methane. With the increasing of the methane/air equivalence ratio, the oxygen component occupying the active center of wall surface reduces, resulting in a low ignition temperature of methane.

(2) Ignition temperature shows differently by the inlet velocity of methane / air mixture. CH₄-Air mixtures inlet velocity which increases limits standing time. Lowering wall surface of Pt(s) involved in the reaction time improves the methane ignition temperature. The active center of Pt(s) component on the wall is gradually increased with inlet velocity.

(3) With the increase of surface catalyst loading density, the area that mixed gas contact catalyst surface per unit time will increase. With the starting wall surface of Catalyst loading density increases, the surface Pt(s) increase correspondingly. This is conducive to the occurrence of primitive reactions $O_2 + 2pt(s) \Rightarrow 2O(s)$. The adding wall catalyst loading density which increases the wall surface of Pt(s) lowers the ignition temperature of methane.

(4) While CH₄-Air mixtures' flow rate keeps constant, the mixed gas pressure improved goes against the oxygen desorption and reduces empty wall surface active center results in the high ignition of methane temperature. At the constant mass flow rate as $PV = \text{const}$, the results show that the ignition temperature basically does not change much with intake pressure at the same mass flow.

(5) With the small equivalence ratio of the methane/air, the small flow velocity, the high wall surface of the catalyst loading density and the intake pressure, this condition is conducive to methane catalytic ignition.

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