



# EFFECT OF WALL THERMAL CONDUCTIVITY ON MICRO-SCALE COMBUSTION CHARACTERISTICS OF HYDROGEN-AIR MIXTURES WITH DETAILED CHEMICAL KINETIC MECHANISMS IN Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> CATALYTIC MICRO-COMBUSTORS

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## ABSTRACT

To understand the effect of different thermal conductivities on catalytic combustion characteristics, effect of thermal conductivity on micro-combustion characteristics of hydrogen-air mixtures in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors were investigated numerically with detailed chemical kinetics mechanisms. Three kinds of wall materials (100, 7.5, and 0.5 W/m·K) were selected to investigate the effect of heat conduction on the catalytic combustion. The simulation results indicate that the catalytic reaction restrains the gas phase reaction in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors. The gas phase reaction restrained by Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalysts is sensitive to thermal boundary condition at the wall. For most conditions, the gas phase reaction cannot be ignored in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors. For low thermal conductivity, the higher temperature gradient on the wall will promote the gas phase reaction shift upstream; high temperature gradient exists on the wall, and the hot spot can cause the material to melt or degrade the catalyst. Due to the gas phase reaction is ignited and sustained in micro-combustors by the heat from the catalytic reaction, the effect of thermal conductivity on micro-scale combustion characteristics is not as obvious as it is in micro-combustors without Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalysts.

**Keywords:** *thermal conductivity; combustion characteristics; catalytic combustion; wall material; micro-scale; Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub>.*

## 1. INTRODUCTION

With the increasing demands on micro devices such as micro-satellite thrusters, micro unmanned aerial vehicles, and chemical micro-sensors and reactors, the needs for the micro-power supply are also increasing, especially on the micro-power source with high-energy density. The alkaline and lithium batteries have quite low specific energy compared with hydrocarbon fuels (Shirsat and Gupta, 2011). Development of micro-combustion based power generating devices, even with relatively inefficient conversions of hydrocarbon fuels to power, would result in reduced weight and increased lifetime of a mechanical or electronic system that currently uses lithium batteries for power. Recent advances in the fields of silicon-based MEMS and silicon micro-fabrication techniques have caused the possibility of the new generation of micro-engines for power generation. Micro-combustor is the critical component for micro-power generation systems using hydrocarbon fuels as the energy source. Several types of chemical micro-reactor and micro-combustor are currently under development (Hua et al., 2005).

In fact, there are some challenges to maintain the stable combustion in micro-combustors. Extinction occurs easily at the micro-scale, because the large area-to-volume ratio increases the heat loss. Another important factor affecting the micro-combustion stability is the quenching distance, which equals to the dimension of micro-combustor (Zhang et al., 2007; Wang et al., 2011).

To improve the micro-combustion stability and the thermal efficiency, various efforts have been paid on the use of catalyst and the optimization of the thermal management, such as the excess enthalpy combustors (Chen et al., 2013b; Chein et al., 2013; Kocich et al., 2012;

Zhong et al., 2012), the heat-recirculating combustors (Belmont and Ellzey, 2014; Chen et al., 2013a; Deshpande and Kumar, 2013; Kurdyumov and Matalon, 2011; Rana et al., 2014; Shirsat and Gupta, 2013), and the catalytic micro-combustors to suppress radical depletion and to enhance the reaction (Federici et al., 2009; Federici and Vlachos, 2011; Hsueh et al., 2010; Hsueh et al., 2011; Wang et al., 2010). Many micro-combustor studies have investigated the variation of dimension, thermal conductivity, inlet velocity, equivalence ratio, etc. (Belmont and Ellzey, 2014; Hua et al., 2005; Ju and Xu, 2005; Raimondeau et al., 2011; Wang et al., 2011; Zhong et al., 2011). Ju and Xu (2005) theoretically and experimentally studied the flame propagation and extinction in micro-channels. Their results showed that the wall thermal properties, flow velocity, and channel width have significant effects on the flame propagation, and cause extinction limits and multiple flame regimes. Leach et al. (2006) and Seyed-Reihani and Jackson (2004) performed a one-dimensional numerically investigated the effects of heat exchange on the reaction zone thickness of stoichiometric premixed hydrogen-air mixtures in micro-channel combustors. They presented an analytical model, which is used to predict the reaction zone thickness of hydrogen-air mixtures based on the thermal properties and the channel size. The heat exchange through the micro-combustors structure can lead to a broadening reaction zone. Vijayan and Gupta (2010 and 2011) performed simulations to highlight the importance of material thermal conductivity and determined the range of thermal conductivities over which combustion could be supported. The results showed that heat loss from the combustion chamber was lower for lower thermal conductivity materials with the overall heat loss being roughly independent of thermal conductivity. However, it is heat lost from the combustion chamber that promotes thermal quenching and not the overall heat loss. Lower thermal

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conductivity materials also have higher rates of heat recirculation to the reactants.

While the micro-combustion has been studied, the effect of the catalytic wall on micro-combustion characteristics are still not fully understood. Most previous computational studies dealt with surface catalytic or gas phase reaction separately and concentrated on the flame stability or extinction limit (Chen et al., 2007; Deutschmann et al., 2012; Ju and Maruta, 2011; Kamijo et al., 2009; Karagiannidis and Mantzaras, 2010; Maruta, 2011). Although the catalyst is used to maintain the reaction and to decrease the heat loss, the effect of thermal conductivity on micro-combustion characteristics and the interaction between surface catalytic and gas phase reaction in micro-combustors are still not fully understood. In this work, micro-combustion characteristics of premixed hydrogen-air mixture in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors are investigated numerically. Numerical simulations with detailed chemical kinetics mechanisms of hydrogen-air mixture combustions in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors are investigated by using the FLUENT coupled with the DETCHEM (Deutschmann et al., 2013). The effect of different reaction models and thermal conductivities on micro-combustion characteristics are discussed.

## 2. NUMERICAL MODELS AND SIMULATION APPROACH

### 2.1 Model geometry and mesh

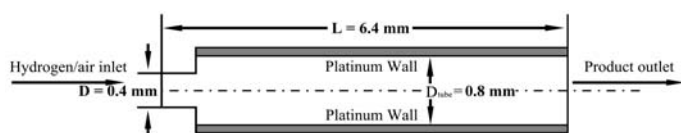


Fig. 1 Schematic diagram of micro-combustor

Table 1 The properties of Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalyst washcoat

| Property  | Value                 |
|---|-----------------------|
| catalyst surface site density $\Gamma$ (mol/cm <sup>2</sup> ) | $2.7 \times 10^{-9}$  |
| average pore diameter $d_{\text{pore}}$ (m)                   | $2.08 \times 10^{-8}$ |
| catalyst porosity $\epsilon_{\text{cat}}$                     | 0.4                   |
| catalyst tortuosity $\tau_{\text{cat}}$                       | 8.0                   |

The geometry of the Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustor is shown in Fig. 1. The micro-tube is modeled with the inner wall coated with Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalyst. The properties of Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalyst washcoat are shown in Table 1. The solid wall material is refractory ceramics (SiC). The aspect ratio of the micro-combustion chamber is kept the same to investigate the effect of dimensional scaling on micro-combustion characteristics. The inlet diameter ( $D$ ), tube diameter ( $D_{\text{tube}}$ ) and tube length ( $L$ ) of the micro-tube are 0.4 mm, 0.8 mm and 6.4 mm, respectively. The wall thickness is 0.1 mm. Premixed; stoichiometric hydrogen-air mixtures is injected into the micro-combustor from the inlet located at one axial end with a step expansion as shown in Fig. 1. The geometry is modeled as a two-dimensional axisymmetric model because of the axial symmetry of the micro-combustor. For all scenarios analyzed, the same grid size of 2  $\mu\text{m}$  is used to mesh the micro-combustion models for the numerical simulations. This fine mesh size will be able to provide good spatial resolution for the distribution of most variables in micro-combustors.

### 2.2 Fluid flow modeling

The reacting gas flow path in micro-combustors and the characteristic length of the combustion chamber, even for power MEMS systems, are still sufficiently larger than the molecular mean-free path of the gases flowing through the systems. Therefore, the fluid media can be reasonably considered as continuum in micro-combustors. The Navier-Stokes equation is solved for the fluid domain and no-slip condition on

the wall is applied. FLUENT 6.3 is used to perform numerical simulations of the fluid flow by solving the conservation equations of energy, momentum, mass, species as well as the conjugated heat transfer condition in the walls. The laminar viscous flow in micro-combustors is considered, and the segregated solution solver of double-precision is applied to solve the above-mentioned set of governing equations. The fluid mixtures thermal conductivity, viscosity, and specific heat are calculated from a mass fraction weighted average of species properties. The fluid density in micro-combustors is calculated using the ideal gas law. In order to couple the heat transfer and the fluid dynamics of the gas-mixtures flow with the detailed chemical kinetics mechanisms (gas phase chemical kinetics and surface catalytic chemical kinetics), an external program DETCHEM 2.5 is applied as user-defined function to FLUENT to extend the modeling capabilities in simulating the detailed chemical kinetics. The DETCHEM software package is designed for modeling and simulation of reactive fluid flows including surface catalytic reaction on catalyst and can apply elementary and multi-step reaction mechanisms in the gas phase and on surfaces (Deutschmann et al., 2013). The CFD simulation convergence is judged upon the residuals of above-mentioned governing equations.

### 2.3 Chemical kinetics mechanisms

Chemical kinetics mechanisms are applied on surfaces and in the gas phase. The surface catalytic reaction mechanism presented by Deutschmann et al. (1996) were employed. The gas phase reaction mechanism consists of 19 reactions and 9 species, which are adopted from the mechanism presented by Miller and Bowman (1989). The above-mentioned chemical kinetics mechanisms have been applied in the previous study (Deutschmann et al., 2000), and the comparisons with experimental results are satisfactory. For hydrogen fuel, five surface species (H(s), O(s), OH(s), H<sub>2</sub>O(s) and Pt(s)) describe the coverage of the surface with adsorbed species. Pt(s) denotes free surface sites which are available for adsorption. The chemical kinetics mechanisms with CHEMKIN format in the gas phase and chemical kinetics mechanisms with surface CHEMKIN on surfaces are imported into the code.

### 2.4 Boundary conditions

The concentration of the hydrogen-air mixtures is specified at the inlet section of micro-combustor. The mass fraction of hydrogen is 0.0283. The inlet temperature and the ambient temperature for the hydrogen-air mixture are 300 K. For all cases studied, a uniform inlet velocity is specified. At the solid wall, the thermal boundary condition is the heat loss to the ambient air. At the outer surface of the solid walls, heat losses to the surroundings are calculated through Eq. (1), in which both natural convection and thermal radiation are considered.

$$q = h(T_{w,o} - T_{\infty}) + \epsilon\delta(T_{w,o}^4 - T_{\infty}^4) \quad (1)$$

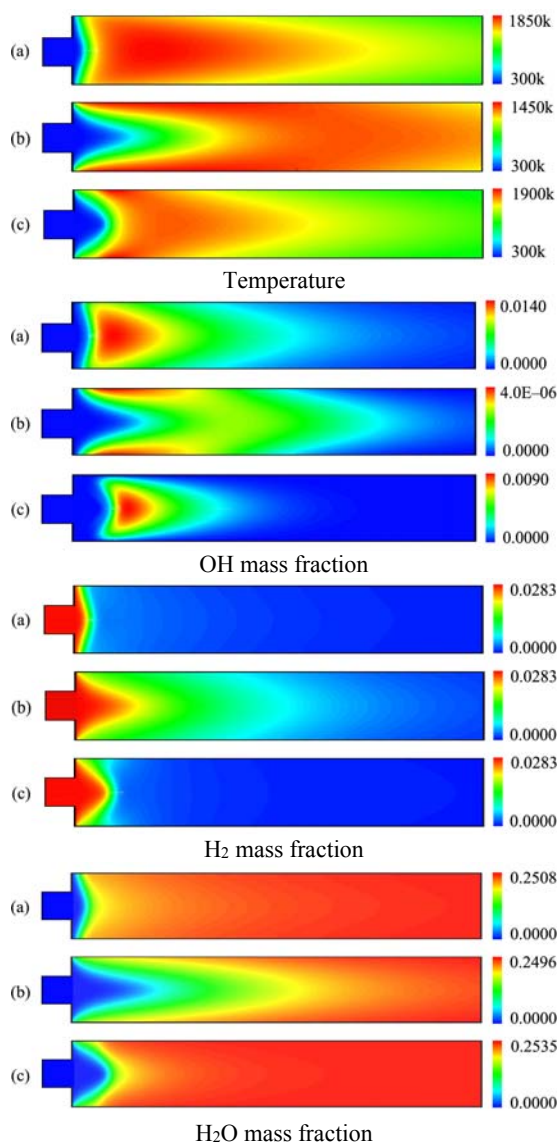
Where the heat transfer coefficient  $h$  is assumed be 20 W/m<sup>2</sup>·K.  $T_{w,o}$  and  $T_{\infty}$  are the outer wall and ambient temperatures, respectively. The emissivity  $\epsilon$  of solid wall is 0.5 and  $\delta$  is the Stefan-Boltzmann constant. At the exit, the far-field boundary condition is not adopted (Norton and Vlachos, 2003), and the pressure is specified at a constant pressure of 0.1 MPa and an extrapolation scheme is used for temperature and species.

## 3. RESULTS AND DISCUSSION

### 3.1 Micro-combustion characteristics for different reaction models

In the present work, the interaction between gas phase reaction and surface catalytic reaction of hydrogen-air in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors is the main issue. In order to clearly identify and to highlight the effect of surface catalytic reaction on micro-combustion characteristics, three different reaction modes by different combinations of the gas phase and surface catalytic reaction mechanism are applied. They are gas phase reaction alone, surface catalytic reaction alone, both

gas phase reaction and surface catalytic reaction mechanisms, respectively.



**Fig. 2** The computed temperature, OH, H<sub>2</sub> and H<sub>2</sub>O mass fraction contours for different reaction models: (a) gas phase reaction alone; (b) surface catalytic reaction alone; (c) both gas phase reaction and surface catalytic reaction

For this case, the wall thickness is ignored. The inlet velocity and diameter of the tube are respectively set to 2 m/s and 0.4mm. The results shown in Fig.2 indicate that the micro-combustion characteristic for different reaction models. In all cases, the comparatively rapid temperature rises close to the inlet was attained, due to highly reactive nature of hydrogen-air mixtures. However, as observed, some differences were seen in these three cases. In the gas phase reaction alone case, the flame structure of micro-combustion displays a cone shape, and the highest temperature and highest OH mass fraction are in the fluid region. Along the radial direction, the temperature is decreased because of the heat is dissipated by the wall. Along the axial direction, the H<sub>2</sub> mass fraction displays a sharp decrease. On the assumption of an inert wall, the OH concentration near the wall reaches a certain value. For the hydrogen and hydrocarbon oxidation, the OH radical is one of significant radicals, and the existence of OH radical generally indicated the high temperature regions and reaction zone. Consequently, OH concentration

is usually used to delineate the gas reaction in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors (Deutschmann et al., 2000). For the surface catalytic reaction alone case, chemical reactions can only occur on the wall. Therefore, the highest temperature is found on the wall near the inlet, and heat transports downstream by convection. The highest temperature is also lower than these for the gas phase reaction alone case or both gas phase reaction and surface catalytic reaction cases. The highest OH concentration also appears on the wall, and its strength is apparently weak since OH radical has high absorption ability. The difference of OH concentration also reveals the variation of surface catalytic reaction and gas phase reaction. For the both gas phase reaction and surface catalytic reaction cases, the high temperature regions exist both in the fluid region and on the wall near the entrance. The high temperature regions are not simply heat convection from the wall, and both gas phase reaction and surface catalytic reaction exist in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors. When compared with the above-mentioned gas phase reaction alone case, the gas phase reaction occurs in the fluid region of micro-combustor, but it shifts slightly downstream. As observed in Fig.2, the gas phase reaction is obviously restrained by the presence of surface catalytic reaction, because most prompt temperature increases are seen in gas phase reaction alone case, not in both gas phase reaction and surface catalytic reaction cases. The above-mentioned tendency can be more evidently seen from the absence of OH radical in the vicinity of inlet, especially in a region close to the Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> wall for both gas phase reaction and surface catalytic reaction cases. For surface catalytic reaction alone case, OH concentration is higher at wall vicinity (maximum value is  $3.96 \times 10^{-6}$ ) than that for both gas phase reaction and surface catalytic reaction cases (maximum value is  $4.77 \times 10^{-8}$ ). More significantly, for surface catalytic reaction alone case, OH radical is only seen in the vicinity of the Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> wall because chemical reaction only occurs on the wall. Product (H<sub>2</sub>O) formation and fuel (H<sub>2</sub>) consumption also indicated that the inhibition of gas phase reaction by Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub>. Note that reaction inhibition by Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> is sensitive to thermal boundary condition at the wall. For most conditions, the simulation results indicate that the gas phase reaction cannot be ignored in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors.

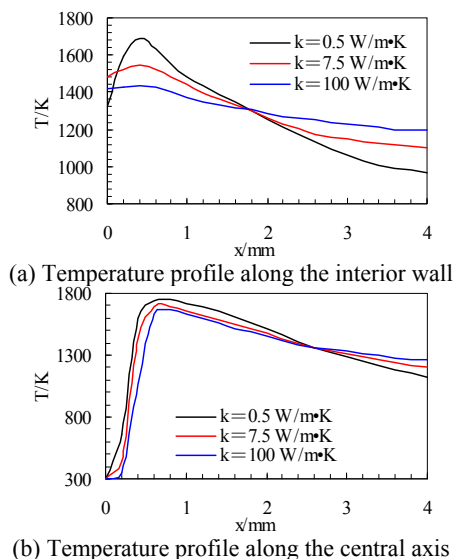
### 3.2 Effect of different wall thermal conductivities

For the materials integrity and flame stability of micro-combustor, the wall thermal conductivity plays a vital role. In micro-combustor, the wall plays a dual, competing role in the overall heat transfer. On one hand, the wall allows exterior heat losses, which can delay ignition and cause extinction. On the other hand, it provides a route for heat transfer from the post-combustion region to upstream for preheating that can improve flame stability and thermal efficiency. The reported simulation results indicated that moderate wall thermal conductivity is essential for flame ignition and stabilization near the entrance of micro-combustor (Norton and Vlachos, 2004).

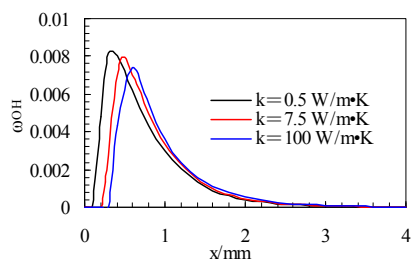
In the present work, the effect of heat conduction within the wall of combustion chamber on micro-combustion characteristics is investigated. Three kinds of wall materials are selected to study the effect of heat conduction on micro-combustion characteristics. First, the low thermal conductivity material such as insulation material is selected. It has relatively low thermal conductivity around 0.5 W/m·K. Second, the medium thermal conductivity material such as ceramic is chosen. Its thermal conductivity is about 7.5 W/m·K. Moreover, the third kind of the selected material has higher thermal conductivity at the level of 100 W/m·K such as metal and silicon. For this case, the wall thickness is 0.1 mm, and the inlet velocity and diameter of the tube are set to 2 m/s and 0.4mm, respectively.

The material thermal conductivity affects the possibility of hot spots and the temperature profile within the wall of combustion chamber. For different material thermal conductivities, the temperature profiles along the interior wall are showed in Fig.3 (a). For lower thermal conductivity materials, hotspot temperatures in excess of 1700 K can occur, an undesirable situation, as it exceeds the maximum operating temperatures of most construction materials. In addition, significant axial temperature gradients are observed in Fig.3 (a). For low wall thermal conductivity, high temperature gradient exists on the wall, and the hot spot can cause

degrade the Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalysts or the material to melt. As the wall thermal conductivity is increased, the wall hot spot is eliminated, and the wall temperature profiles become more uniform. Despite the apparent advantages of higher wall thermal conductivity for material stability, most materials that offer high thermal conductivity are metals, and therefore would not be inert to radical quenching. A more reasonable solution would be thicker walls of a more inert material that may have a lower thermal conductivity.



**Fig. 3** Temperature profile along the interior wall and central axis for different wall thermal conductivities.



**Fig.4** OH mass fraction profile along the central axis for different wall thermal conductivities

The temperature distribution along the central axis for different wall thermal conductivities is shown in Fig.3 (b). As observed, it shows a higher temperature for low wall thermal conductivity. The OH distribution along the central axis for different wall thermal conductivities is shown in Fig.4. For high thermal wall conductivity, the flame location shifts slightly downstream, and a lower OH concentration is found in combustion chamber. A high temperature gradient on the wall will make the gas phase combustion shift upstream, and the combustion chamber will have a higher peak temperature. However, the above-mentioned behavior is different from that in micro-combustors without Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalyst walls. In that case (Hua et al., 2005), as the wall thermal conductivity is decreased, the flame core in combustion chamber will shift downstream. The simulation results indicate that the effect of wall thermal conductivity on micro-combustion characteristics is not as evident as that in micro-combustors without Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalyst walls. The effect of different wall thermal conductivities on micro-combustion characteristics is not so obvious, because the heat to ignite the gas phase reaction is primarily from the surface catalytic reaction on the wall, not from the upstream heat conduction.

## 4. CONCLUSIONS

In this study, micro-combustion characteristics for different combustion models and the effects of different wall thermal conductivities on surface catalytic combustion of hydrogen-air mixtures in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors were investigated numerically with detailed chemical kinetics mechanisms. From this study, the following conclusions are obtained.

For most conditions, the gas phase combustion cannot be ignored in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors. The surface catalytic combustion restrains the gas phase combustion in Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalytic micro-combustors. The gas phase combustion inhibition by Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalysts is sensitive to thermal boundary condition at the wall.

For low wall thermal conductivity, the higher temperature gradient within the wall of combustion chamber will promote the gas phase combustion shift upstream, and will result in higher temperature distribution in combustion chamber. The effect of different wall thermal conductivities on micro-combustion characteristics is not as obvious as it is in micro-combustors without Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalyst walls, because the gas phase combustion is ignited and sustained by the heat from the surface catalytic combustion.

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