A Study on Premixed Catalytic Combustion of Propane*

Feng YE**, Tsuyoshi NAKAJIMA***
and Yuji IKEDA***

The Catalytic combustion of a lean premixed propane mixture was investigated below the catalyst temperature of 650°C and without gas-phase reaction, using a platinum-impregnated foam catalyst. The temperature distribution in the catalyst plate and the composition of the exhaust gas were measured at air ratios of about 2.2 to 5.8 and heat loads 43 kW/m² to 115 kW/m² under stable combustion conditions. Since the maximum temperature was observed very near the inlet of the catalyst plate, it is thought that the reaction rate of the catalyst is high and the reaction zone is narrow. When the air ratio was decreased while keeping the heat load constant, the maximum temperature in the catalyst plate increased, but the outlet surface temperature of the catalyst plate was almost constant. The temperature distribution of the outlet surface of the catalyst plate was influenced by natural convection and showed higher temperature in the upper region. The combustion efficiency was strongly related with the catalyst temperature and showed almost the same value when the maximum temperatures are the same. The range of stable catalytic combustion was investigated and its relation to the catalyst thickness was determined.

Key Words: Catalyst, Catalytic Combustion, Premixed Combustion, Gaseous Fuel, Lean Burn, Catalyst Temperature, Combustion Efficiency, Blowout

1. Introduction

The application of catalytic combustion by surface reaction to medium- or low-temperature heating devices, such as domestic and industrial heaters, has been eagerly awaited from the perspective of cleanliness and safety in recent years^{(1)–(3)}, since catalytic combustion is a stable and highly efficient self-sustained process even when the combustion temperature is low and the premixed gas is very lean, compared to gas-phase combustion (i.e., flame combustion).

Most of the recent studies on catalytic combustion devices are conducted at high load and high velocity, with combustion temperatures above 700°C. Studies on the catalytic combustion at high temperature involve not only the combustion phenomenon on

the catalytic surface but also the influence of gas-phase combustion reaction^{(4),(5)}.

Other researchers, taking into consideration safety at low-temperature catalytic combustion in practical use seriously, have developed flameless catalytic combustion burners which are efficient at catalytic combustion temperatures below 700°C, using a porous ceramic fiber combustion catalyst. The combustion method employed in these catalytic combustion burners is the diffusion combustion method which causes combustion in the catalytic layer by diffusion of air indispensable for combustion from the front of the catalytic mat and provision of fuel from the inside of the mat. With regard to this method, however, a problem has been reported that, when the heat load is markedly increased, the combustion efficiency declines due to lack of air caused by diffusion velocity reaching its limit^{(3),(6),(7)}.

The premixed catalytic combustion method, in which air and fuel are mixed in advance, is expected to overcome this problem and easily control the combustion conditions such as combustion temperature and exhaust gas characteristics, because combustion can be achieved with the air ratio and the heat load

* Received 18th October, 1993, Japanese original: Trans. Jpn. Soc. Mech. Eng., Vol. 59, No. 558, B (1993), p. 613-618 (Received 17th July, 1992)

** Graduate School of Natural Science and Technology, Kobe University, 1 Rokkodai, Nada-ku, Kobe 657, Japan

*** Department of Mechanical Engineering, Kobe University, 1 Rokkodai, Nada-ku, Kobe 657, Japan

set in advance. Despite these advantages, few studies have been conducted on the premixed catalytic combustion method featuring low-temperature combustion with little gas-phase combustion reaction. It is, therefore, desired to reveal the behavior of catalytic combustion and its reaction mechanism, and evaluate its benefits.

The present study examined catalytic heat characteristics, combustion characteristics and combustion range by observing the temperature distribution and measuring the components of exhaust gas in the catalytic layer, when the premixed catalytic combustion of propane fuel was conducted using a foam catalyst below 650°C, i.e., the heat-resisting limit of the catalyst under lean ratio below the gas-phase combustion limit.

Symbols

λ : air ratio

T : temperature, °C

$C_{C_3H_8}$: mole fraction of propane, mol%

V_0 : velocity in the standard state, cm/s

q : heat load, kW/m²

η : combustion efficiency, %

δ_c : thickness of catalytic mat, mm

X : coordinate in the flow direction from the catalyst inlet, mm

Y : coordinate in the vertical direction from the center of the catalytic mat, mm

Subscripts

c : catalyst

max : maximum

in : inlet of catalytic mat

out : outlet of catalytic mat

UHC : unburnt hydrocarbon

2. Experimental Apparatus and Combustion Procedure

2.1 Experimental apparatus

Figure 1 presents the experimental apparatus and measurement system which consist of fuel and air supply systems, premixing part, catalytic combustion section, temperature measuring system, and sampling system for gas concentration. As shown in Fig. 1, the catalytic combustion section consists of the catalytic layer, the electric heater, and the heat-resistant layer. The catalytic layer 9 mm or 18 mm thick is used by stacking three or six 3-mm-thick 17 cm square sheets. The wall surrounding the catalytic layer is insulated with 50 mm-thick spongy heat-resistant layer in order to prevent heat loss. The catalyst is a platinum-impregnated porous foam catalyst made by Nikki Universal Co., Ltd. Physical properties of the catalyst are listed in Table 1. The propane of 97% purity is

used as fuel.

2.2 Experimental procedure

Propane is supplied directly from the gas pressure vessel. It is drawn into the mixing pipe through a nozzle after its flow quantity is measured with a flow meter, and mixed with air supplied by the blower and measured with a flow meter. The air-fuel mixture passes through the divergent zone and enters the catalytic layer after being straightened through the metallic mesh and the porous insulator. At the beginning of combustion, the catalytic layer is electrically heated with a nichrome heater, maintaining the quantity of air flow at a predetermined rate. When the fuel is supplied at a given flow rate after the temperature of the catalytic layer reaches that (approx. 220°C) at which the catalyst begins to oxidize propane, the catalytic combustion starts and the temperature rises abruptly. Then the electric heating is terminated. After the temperature of catalytic layer is gradually stabilized by the catalytic combustion, the temperature of the catalyst and the components of exhaust gas are measured.

Temperature measurement in the catalytic layer is conducted by inserting a thermocouple connected with the traverse unit into a 4-mm-diameter hole at the back surface of the catalytic layer. Since the temperature measured by this method is almost the same as that obtained by the thermocouple originally embedded in the catalytic layer without holes, the existence of such a hole seems to have little influence on the combustion in the catalytic layer.

Table 1 Physical properties of catalyst

Geometric Surface	10 ³ m ² /m ³
Space Ratio	93 %
True Density	8.6 g/ml
Bulk Density	0.6 g/ml
Pt Supported Quantity	1.0 g/l
Pt Particle Size	~10 Å

Fig. 1 Experimental apparatus

Combustion gas is sampled at the exhaust shown in Fig. 1. Concentrations of various hydrocarbons such as C_3H_8 , $i-C_4H_{10}$, $n-C_4H_{10}$, C_2H_6 are measured by gaschromatography equipped with TCD and FID detectors. Combustion efficiency is calculated from the following formula derived from the chemical equation for a one-step reaction of C_3H_8 , with the known air ratio of the mixed gas.

$$\eta = \frac{23.81 \times C_{UHC} \times \lambda - 1 + C_{UHC}}{3 \times C_{UHC} - 1} \times 100$$

Since the fuel used in the experiment is propane (C_3H_8) with 97% purity and the main unburnt substance detected in the combustion exhaust gas is C_3H_8 , the result from the above formula of combustion efficiency is likely to have little error in calculation even if the concentration of the unburnt substance, (C_{UHC}), is calculated by converting hydrocarbon components in unburnt substances into C_3H_8 .

3. Experimental Result and Considerations

3.1 Influence of air ratio and heat load on temperature distribution in the catalytic layer

Figure 2 shows temperature distributions from the upstream surface to the downstream surface along the central axis of the catalyst, with the air ratios of 3.8 and 4.6 and the heat loads of 57 and 86 kW/m^2 . (Temperature measuring points, unless otherwise noted, all lie along the central axis of the catalyst.) As shown in Fig. 2, when the air ratio is fixed, the catalyst temperature always increases as the heat load (fuel flow rate) increases. The catalyst temperature generally increases abruptly from its inlet and decreases gradually after reaching the maximum temperature. Since the maximum temperature is observed near the catalyst inlet, the reaction rate of the catalyst seems to be very high and the reaction zone seems to be narrow.

Figure 2 also reveals that when the heat load and the air ratio are low, i.e., when the flow rate of the mixed gas is low, the maximum temperature is located almost at the inlet surface; however, it gradually deviates from the inlet surface as the flow rate of the mixed gas increases with increases of air ratio and heat load. The catalytic inlet temperature decreases with the increase of the mixed gas flow rate, which can be considered that the cold mixed gas cools down the catalyst inlet when it flows into the catalyst, and the resultant low temperature around the inlet deactivates the combustion reaction at the catalyst surface.

Figure 3 shows the catalytic temperature distribution when the air ratio is varied under a constant heat load. Although, near the inlet, the catalyst temperature increases with decreasing air ratio, little

difference is observed among the three air ratios as the temperature measuring point deviates from the inlet. The high temperature in the front half of the catalyst is considered to be caused by two reasons: one is that because of low air ratio, i.e., high-concentration mixture, the heat-generation rate increases

Fig. 2 Influence of heat load on temperature profiles

Fig. 3 Influence of air ratio on temperature profiles

with the increase of fuel diffusion into the catalyst surface, and the other is that the heat transfer from the catalyst to the gas deteriorates with low gas velocity. This rise of the catalyst temperature increases the reaction rate on the catalyst surface, so that the maximum temperature point in the catalyst, as shown in Fig. 3, moves to the upstream surface with the decrease of air ratio and the combustion zone becomes narrower.

As shown in Fig. 3, the catalytic inlet temperature varies markedly with the change of air ratio, while little change can be seen in the outlet temperature. Figures 4 and 5 show the catalytic inlet and outlet temperatures at various air ratios when using heat load as a parameter. The catalytic inlet temperature decreases markedly with the increase of air ratio, and its rate of decrease becomes greater as the heat load increases. In contrast, as shown in Fig. 5, the outlet temperature increases as the heat load increases. It is, however, influenced very little by the change of the air ratio, and becomes constant when the heat load is fixed.

Fig. 4 Influence of air ratio and heat load on catalytic inlet temperature

Fig. 5 Influence of air ratio and heat load on catalytic outlet temperature

For utilization of heat produced by low-temperature catalytic combustion, infrared rays from the catalytic surface are confirmed to be effective. For this purpose, influence of some combustion conditions of the mixture, i.e., air ratio and heat load, on the temperature distribution of the combustion catalyst should be examined. From the above-mentioned results, the outlet surface temperature of the catalyst in the premixed catalytic combustion does not depend on the air ratio in the stable combustion zone, rather it only increases when the heat load (i.e., amount of fuel) increases. Because of this, the radiative heat from the outlet surface can be easily controlled. This seems to provide many benefits for practical use.

3.2 Temperature distribution of the catalytic outlet surface

By measuring the outlet surface temperature of the catalyst plate, the following are revealed: in the horizontal direction, in most of the central region of the catalyst, the temperature is distributed symmetrically about the central axis, while in the vertical direction, the temperature gradually increases toward the upper region. These may be influenced by natural convection resulting from the use of the catalytic mat set in the vertical direction. Since the periphery temperature of the catalyst was much lower than the temperature at its central part, thermal insulation can be regarded as insufficient. Figures 6 and 7 show the results of the temperature measurements in the vertical direction along the central axis of the catalytic outlet surface. In most of the central region of the catalyst, the temperature change is slight; however, it is large near both upper and lower ends of the vertical catalytic plate. These figures also show that since the influence of natural convection on the catalytic combustion increases in the low heat load region with relatively low air ratio, i.e., low velocity of mixed gas flow, the temperature change in the vertical

Fig. 6 Influence of air ratio on surface temperature profiles at the catalytic outlet

direction becomes large. In addition, to examine the influence of natural convection on the temperature distribution in the catalytic layer, the catalyst temperature was measured at various points 5 cm below the central axis of the catalyst and compared with that measured along the central axis in Fig. 8. The temperature distribution at the points 5 cm below is influenced by natural convection and shows in general, values ten degrees lower than those measured at the center.

The catalytic combustion under low heat load and low temperature has the following advantages: uniformity of the surface temperature of the combustion catalyst and large heat radiation from the surface. In light of these, its application to domestic heaters and dryers is expected. However, it has also been confirmed that there is a temperature at which the oxidation reaction of the catalyst is initiated in the catalytic surface combustion, and the combustion reaction rate is closely related with the catalytic

temperature. Hence, from the present experiment, it is considered that the temperature distribution of the combustion catalyst is not uniform: the periphery temperatures of the catalyst become lower than those at the center due to heat loss, resulting in the deterioration of average combustion efficiency. In light of this, much attention should be paid to determining the best methods for installation of the catalyst and achieving good heat insulation.

3.3 Blowout and stable combustion zone

As shown in Fig. 3, under the constant heat load, the catalytic inlet temperature and the maximum catalytic temperature decrease with increase of air ratio, i.e., increase of the mixed gas quantity, and the location showing the maximum catalytic temperature moves downstream. When the air ratio is increased, the maximum catalytic temperature decreases, and the catalytic combustion begins to show instability as the temperature approaches 400°C. With passage of time, the temperature decreases slowly and finally the combustion terminates. This is shown on the monitor display in Fig. 1. The reason why stable combustion does not last and blowout appears may be that the heat transfer caused by conduction and convection in the catalytic combustion exceeds the heating value by the chemical reaction, thus destroying the heat balance.

As mentioned earlier, under the constant heat load, although the maximum catalytic temperature increases as the air ratio decreases, it is of concern that the temperature might exceed the catalytic heat endurance limit, i.e., 650°C for the catalyst used in this experiment. On the other hand, when the air ratio increases, the blowout appears at a certain air ratio. Considering these limits, the catalytic temperature were measured in a wide range of heat load values to determine the stable catalytic combustion zone. The results of measurement are shown in Fig. 9. Since blowout occurs at lower air ratio and the plot of limit temperature (650°C) shifts to the higher air ratio with increasing heat load, the range of the air ratio under which the catalytic combustion is stable gradually becomes narrower and the maximum velocity of the mixed gas flow is limited. This is because the heat transfer quantity from the catalytic surface to the gas increases and the combustion heat balance is destroyed with increase of the flow velocity of the mixed gas. This result agrees with that of work by Sato and Sugimoto⁽⁸⁾.

The combustion efficiency calculated from the components of the exhaust gas is also shown in Fig. 9. The efficiency is strongly related with the maximum combustion temperature. As shown in Fig. 9, the contour lines of the combustion efficiency (chain

Fig. 7 Influence of heat load on surface temperature profiles at the catalytic outlet

Fig. 8 Comparison of catalytic temperature profile measured 5 cm below the central axis with that at the central axis

Fig. 9 Combustion range for 18-mm-thick catalyst

lines) and the maximum combustion temperature (solid lines) show the same tendency. Even when the maximum catalyst temperature is relatively low, i.e., 550°C, the combustion efficiency reaches 97%.

It is reported for the catalytic diffusive combustion that the range of heat load is narrow because of the low intake air quantity and the largest heat load is about 25 kW/m²⁽⁷⁾. The test apparatus of catalytic premixed combustion used in this experiment can produce the heat load 5 times as large as that of the catalytic diffusive combustion at the same catalytic temperature as shown in Fig. 9.

From the above-mentioned results, in the premixed catalytic combustion method, the combustion efficiency and the catalytic temperature, in particular the catalyst outlet surface temperature related to the radiative heat can be adjusted to meet the actual demand, and combustion can be more easily controlled than the catalytic diffusive combustion method.

3.4 Influence of catalyst thickness on the combustion range

All of the above-mentioned results are obtained using a catalyst whose thickness is 18 mm. The combustion range and the contour lines of the maximum catalyst temperature and combustion efficiency are shown in Fig. 10 when the catalyst thickness is decreased to 9 mm. We see that the blowout line is much lower than that of the 18-mm-thick catalyst, which is within the air ratio range below the catalytic limit temperature line of 650°C, and the stable combustion range surrounded by those lines becomes considerably narrower than that using the 18-mm-thick catalyst. Figure 10 also shows that the combustion

Fig. 10 Combustion range for 9-mm-thick catalyst

efficiency is inferior to that of 18-mm-thick catalyst at the same maximum catalyst temperature. Using the 9-mm-thick catalyst, under the same conditions of the mixed gas, since the average contact time of the mixed gas with the catalytic surface becomes shorter, the surface reaction is not enough due to the insufficient time, and the heat loss increases because the catalytic radiation area per unit volume is greater than that of the 18-mm-thick catalyst. These are considered to be the reasons why the stable combustion range becomes narrower. The most appropriate catalyst thickness for the catalytic combustion should be carefully examined in future studies.

4. Conclusion

An experiment of catalytic combustion at low temperatures was carried out using a platinum-impregnated metallic porous foam catalyst for lean premixed propane beyond the limit of gas-phase combustion concentration, and limiting the highest catalytic temperature to 650°C in order to examine the influence of air ratio, heat load, and natural convection on the temperature distribution of the catalytic layer, the stable catalytic combustion range and the combustion efficiency. The following results are obtained.

(1) When the combustion is stable, catalytic reaction starts immediately and the combustion reaction zone is narrow, since the maximum temperature point of the catalytic layer is near the inlet.

(2) The temperature in the catalytic layer increases with increase of heat load. Under the constant heat load, the maximum catalyst temperature increases with decrease of the air ratio. Since the

catalyst outlet temperature changes little, and the radiative heat intensity from the outlet surface can be controlled by the heat load quantity, these will be beneficial to practical use.

(3) When putting the catalyst vertical, the catalytic combustion is influenced by natural convection and results in the temperature gradient in the vertical direction of the catalyst. The influence of natural convection is significant under a low premixed gas velocity.

(4) Catalyst combustion efficiency is strongly related with the maximum catalyst temperature. The combustion efficiency showed almost the same value when the maximum catalyst temperatures were the same.

(5) In the catalytic combustion range surrounded by the maximum temperature line determined by the catalyst durability and the combustion blowout line, the air ratio range becomes gradually narrower with increase of the heat load quantity, and finally the maximum processing flow rate of the gas is determined. When the catalyst thickness is decreased from 18 mm to 9 mm, the stable combustion range becomes narrower.

Acknowledgment

We thank Kagura Invest. Co., Ltd., and Mr. Akira Michiwaki of Kobe University for his technical assistance in this experiment.

References

- (1) Haruta, K., Fuel and Combustion, (in Japanese), Vol. 45, No. 5 (1978), p. 385.
- (2) Nishino, A., Suzuki, T., Takeuchi, Y. and Ono, Y., J. of the Fuel Soc. of Japan, (in Japanese), Vol. 66, No. 4 (1987), p. 287.
- (3) Dongworth, M.R. and Melvin, A., Proc. 16th Symp. (Int.) Combust., (1976), p. 255.
- (4) Mori, Y., Miyauchi, T. and Danzuka, T., Trans. Jpn. Soc. Mech. Eng., (in Japanese), Vol. 49, No. 445, B(1983), p. 1973.
- (5) Nomura, Y., Ohkubo, Y. and Ohsawa, K., Trans. Jpn. Soc. Mech. Eng., (in Japanese), Vol. 57, No. 534, B(1991), p. 710.
- (6) Trimm, D.L. and Lam, C.-W., Chem. Eng. Sci., Vol. 35, (1980), p. 1731.
- (7) Sadamori, H., Ito, S. and Okada, O., J. of the Fuel Soc. of Japan, (in Japanese), Vol. 67, No. 12 (1988), p. 1052.
- (8) Sato, M. and Sugimoto, T., Proc. 28th National Combust. Symp. of Japan, (in Japanese), (1990), p. 560.