

STUDY ON THE CHARACTERISTICS OF CATALYTIC COMBUSTION FURNACE OF NATURAL GAS AND INFLUENCE OF ITS EXHAUST GAS TO PLANT

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ABSTRACT

This article discussed the radiation and pollutant emissions characteristics of the catalytic combustion furnace based the combustion of lean natural gas-air mixtures in catalytic honeycomb monoliths and the influence of its exhaust gas on schefflera plants growth by means of theory and experiments. The radiation efficiency of the monolith alone varied from approximately 20% to 40%. The glazed tiles heated by the catalytic combustion furnace are more fine and glossy than that of conventional ones. Schefflera plants in experimental group in a greenhouse filling with catalytic combustion exhaust gas. On contrary, schefflera plants in control group stay in indoor environment. The result showed that schefflera plants in experimental group grow faster than that in control group, even greener. The content of pollutants in the exhaust gas of furnace and the date of the parameter of temperature, humidity and the concentration of CO₂ in greenhouse were measured. Catalytic combustion as a developing technology could make the pollutant emissions (CO and NO_x) to near zero. The exhaust gas with high CO₂ concentration could promote the growth of plants.

Keywords: *Catalytic combustion furnace; Radiation; CO₂ concentration; Plant.*

1. INTRODUCTION

Air pollution is a critical environmental issue for the world, which is also a global problem with far-reaching environmental impacts. These mainly related to the traffic sector, but also regarding the industrial and residential combustion sectors (Duque *et al.*, 2016). Recent epidemiologic studies have shown that ambient air pollution exposure is associated with increased mortality and higher incidence diseases such as asthma, chronic obstructive pulmonary disease, and cancer (Cohen *et al.*, 2005). When the first scientist examined the effect of air pollution on health after the Meuse Valley fog in 1930 or the London smog in 1952, no one could have known what an extended field of research they started (Rückerl *et al.*, 2011).

Because of the great interest in a process which both reduced combustion pollutant emissions and also improved thermal efficiency, many research programs resulted (Pfefferle and Pfefferle, 1987). Catalytic combustion has been known since 1818, Sir Humphrey Davies observed that coal-gas and oxygen were able to sustain a combustion reaction on a platinum wire in the absence of a flame, giving off thermal radiation from the wire in its place. Heterogeneously catalyzed gas-phase combustion or catalytically stabilized combustion was first demonstrated by Pfefferle in 1970 (Pfefferle, 1975) and was shown to be an efficient method for burning fuels in lean fuel-air mixtures without significant formation of pollutants (Pfefferle, 1975; Pfefferle, 1978; Zhang *et al.*, 2010).

The combustion of hydrocarbons contributes to a large part of air pollutant and the formulation of particulate matter (PM), however, catalytic devices for complete low-temperature oxidation of hydrocarbons were employed in various areas in place of traditional

flaring devices and had the following advantages over the conventional heat sources: complete combustion of the fuel, which enhanced combustion efficiency; lower combustion temperature (Kirillov *et al.*, 2003).

At present, the research of catalytic combustion technology was mainly related to the preparation of the catalyst, while the research of the application of catalytic combustion technology was few relatively. Recently, Anna-Karin Jannasch (Jannasch *et al.*, 2006) constructed and evaluated a new Catalytic burner prototype, based on Catator's patented wire mesh catalysts, for natural gas combustion in gas cooking stoves. The results were very promising with respect to the catalyst life-time (>10 000 h), the NO_x and the CO-emissions was low. In addition, a relatively high thermal efficiency could be measured over abroad range of power inputs (around 40–50% for 1–4 kW). The catalytic combustion of CH₄/air mixtures over Pd-based catalysts was investigated experimentally and numerically at gas turbine relevant conditions (pressures up to 15bar and inlet temperatures up to 723K) in a reactor comprising of alternately coated channels. A global catalytic reaction rate was deduced, valid for catalyst temperatures below the PdO decomposition temperature (Carroni *et al.*, 2003). Fibers obtained using the PVB/ cerium nitrate reached higher conversions of methane, indicating higher catalytic activity. The electrospun fiber produced small amounts of nitrogen compounds during the catalytic combustion of methane (Pontelli *et al.*, 2011). In the present study the radiation and pollutant emissions characteristics of the catalytic combustion furnace were concerned. To better understand the environmental effects of catalytic combustion, influence of its exhaust gas to plant was also conducted.

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2. EXPERIMENTAL SET-UP

Figure 1 illustrates the system diagram of catalytic combustion furnace. The furnace body had the shape of a cuboid, whose edges were 500 mm long, 500 mm wide and 660 mm high. It was connected to the catalytic burner. The support for all the monoliths tested here was cordierite and its softening temperature was 1380 °C. The two square catalytic honeycomb monoliths were installed in the burner each time. The square honeycomb monoliths were 150 mm wide and 20 mm long, with square-shaped cells which sectional area was 1mm×1mm. In order to decrease the temperature of mixtures in chamber connected with the monolith's entrance, the 20mm long blank monoliths were inserted between the chamber and the Pd based catalytic monolith's entrance as assembly of monolith.

The reactant gas feeds of natural gas and air were regulated via GMS0050BSRN200000 natural gas meter and CMG400A080100000 air meter with 0~50 L/min and 0~80 m³/h of full-scale ranges, respectively. The two meters were provided by electric current. The cooling circulating water was used to cool the burner chamber.

The sampling line of exhaust gas from furnace vent consisted of a probe, taking an average exhaust sample, followed by tubing. Exhaust gas went into the greenhouse in which the schefflera plant was settled in. To supplement the data, mixed gas in the greenhouse also was analyzed by the exhaust gas analyser and the air parameter was recorded by digital thermo-hyrometer, offering the information of ambience which plant was in. During the experiment, glazed tiles in furnace had still been heated. First of all, we divided four same growth statement schefflera plants into two groups, experimental group and control group. Schefflera plant in experimental group stay in a greenhouse filling with catalytic combustion exhaust gas, on contrary, schefflera plant in control group stay in general environment. All plants watered equally and kept recording their statement should be made sure.

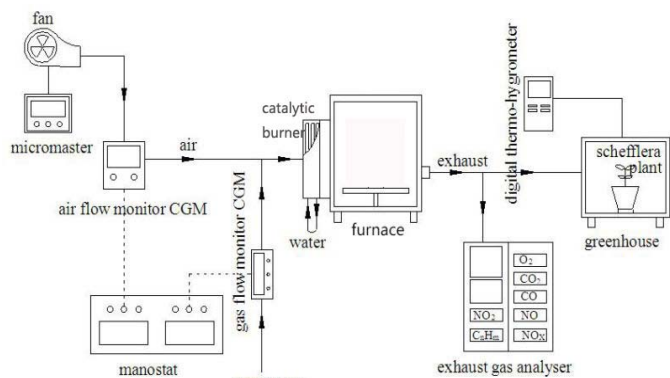


Fig. 1 The system diagram of catalytic combustion furnace

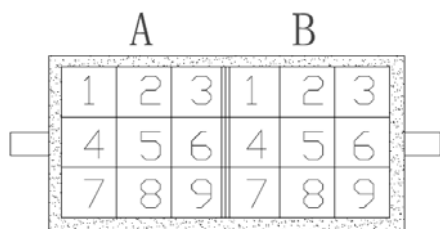


Fig. 2 Surface of the two monoliths

As shown in Fig. 2, the surface of the two monoliths was divided into eighteen uniform regions with the sectional area of 50mm×50mm. The local surface temperature of catalytic monolith was measured by UX-20P infrared radiation thermometer with 600~3000 °C of full-scale ranges. And the exhaust gas temperature at open end of monolith was measured by type thermocouple K of diameter 0.5.

3. EXPERIMENTAL RESULT AND DISCUSSION

3.1 The radiation outputs of the monoliths

Figure 3 plots average local surface temperature of monolith and average gas temperature at the open end of monolith between 4 and 13.09 kW in thermal input of catalytic burner. As we can see from the profiles, average local surface temperature and average gas temperature ascended gradually with the increasing thermal input firstly, then their temperature began to decrease when the thermal input reached 12 kW. The combustion was stable between 4 kW and 10 kW. But a part area of catalyst became black gradually at 13.09 kW.

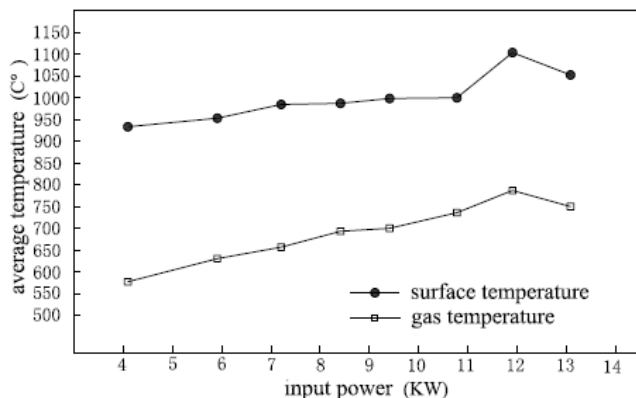


Fig. 3 Curves of average local surface temperature of monolith and average gas temperature at open end of monolith

The radiation outputs of the monoliths studied here were calculated based on the surface temperatures measured by the infrared pyrometer and using an effective emissivity ϵ_{eff} of 0.5. Equation (1) was used:

$$\begin{aligned} \text{Radiation Efficiency (\%)} &= 100 \times \frac{\text{radiation output}}{\text{thermal input}} \\ &= 100 \times \frac{FA \epsilon_{eff} \sigma (T_s^4 - T_a^4)}{\dot{V}_{CH_4} \times CV_{CH_4,net}} \end{aligned} \quad (1)$$

where F is a view factor of around 0.6 which accounts for a section of the burner with square honeycomb monoliths. A is the apparent area of the monolith at the open end, σ is the Stephan-Boltzmann constant, T_s is the average surface temperature and T_a is the temperature of the surroundings. \dot{V}_{CH_4} is the volume flow rate of CH₄ at STP and $CV_{CH_4,net}$ is the net calorific value of CH₄ at STP (Dupont *et al.*, 2000). The main composition of natural gas is methane. Here, the net calorific value of natural gas is 34.54 MJ/Nm³.

Figure 4 shows the radiation efficiency of the monolith alone between 4 and 13.09 kW in thermal input of catalytic burner.

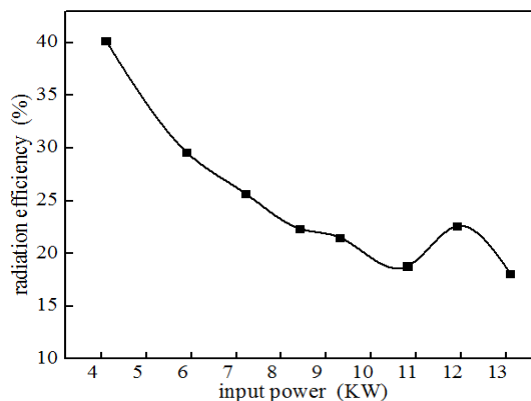


Fig. 4 Curves of the radiation efficiency in different thermal input of catalytic burner

The radiation efficiency of the monolith alone varied from approximately 40 % to 20 % (for total thermal inputs of 4 kW and 10 kW respectively). As the thermal inputs increases, there has been a gradual drop in the radiation efficiency of the monolith alone up till 10 kW. After that, the catalytic combustion of burner was not very steady.

3.2 Radiation heat transfer of the catalytic combustion furnace

When reaching a steady state of catalytic combustion, the monolith was a bright red. Radiation heat exchange between the surface of catalytic honeycomb monoliths and the wall surface in an enclosed furnace was regarded as the radiation heat exchange between two gray bodies in an enclosed cavity. It was calculated by Eq. (2):

$$\phi_{1,2} = \frac{A_1 \sigma (T_1^4 - T_2^4)}{\left(\frac{1}{\epsilon_1} - 1\right) + \frac{1}{X_{1,2}} + \frac{A_1}{A_2} \left(\frac{1}{\epsilon_2} - 1\right)} \quad (2)$$

where ϵ_1 is the emissivity of catalytic honeycomb monoliths surface, which was 0.5, the wall surface was surrounded by heat sink, which ϵ_2 was 0.85, $X_{1,2}$ is the shape factors, here was 1, A_1 is the apparent area of the monolith at the open end, which was $A_1 = 0.15 \times 0.3 = 0.045 \text{ m}^2$. The inner casing of furnace still had the shape of a cuboid, whose edges were 370 mm long, 150 mm wide and 300 mm high. A_2 is the apparent area of the wall surface, which is $A_2 = 0.378 \text{ m}^2$. T_1 is the average surface temperature of monoliths, which was 1373 K (Zhang *et al.*, 2008). T_2 is the average temperature of the wall surface, which was recorded approximately as 1173 K by thermocouple K of diameter 1. At this moment natural gas flow rate was 8.5 L/min (input power of 4.893 kW), but partial thermal input was waste to the surroundings due to the construct of furnace system. The furnace here was supposed to be as an adiabatic system. Thus the radiation heat transfer of the catalytic combustion furnace is calculated to be 2.097 kW. The radiation efficiency of the monolith alone is difference from that of the monolith in an enclosed furnace. Because an enclosed furnace which surrounds the monolith's output in radiation could reflect or re-emit it entirely to heat sink in the furnace. So the values of the radiation of the monolith in an enclosed furnace is higher than that in figure 4. This high radiant heat exchange inside furnace had a key role to heating glazed tiles.

Figure 5 shows the glazed tile heated by the catalytic combustion furnace.



Fig. 5 Glazed tile heated by the catalytic combustion furnace

The glazed tiles possessed excellent color and luster of glaze surface. Overall, the glaze surface of the tile was very smooth, and it created a strong artistic atmosphere.

3.3 Pollutant emissions of the catalytic combustion furnace

Figure 6 plots exhaust gas pollutant emissions at exhaust sampling point of catalytic combustion furnace vent as time went on. At the time of the ignition we recorded it as zero at the same point which was connected with pipes of analysers.

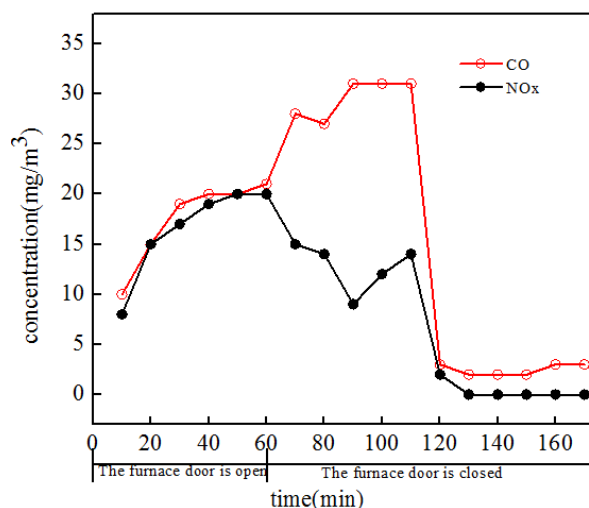


Fig. 6 curves of emissions in the exhaust gas of catalytic combustion furnace

It was shown that NOx rose during the first period of 60 minutes due to the appeared flame influence of gas combustion. Then NOx decreased between 60 and 170 minutes. But the emission of NOx was low (below 20 mg/m³) because the exhaust gas had been significantly diluted before the furnace door closed. Then the catalytic combustion entered a steady state gradually with the decreasing NOx after 60 minutes. NOx dropped to near zero when regulated the flow rate of natural gas to 7.0 L/min at 120 minutes, because the temperature is lower than that of the generation of NOx and the oxidation reaction of catalytic combustion is almost complete.

The emission of CO ascended gradually as time went on during the first period of 110 minutes. The maximum value of CO is 31 mg/m³. It was clear that the emission of CO was increasing faster after 60 minutes. Because there were CO selectivities for the fuel-rich concentrations (conditions of partial oxidation) during the start-up process. And CO was not diluted during the furnace door closed after 60 minutes. Similarly, when the burner reached a steady catalytic combustion state in the condition of lean-fuel, CO dropped to near zero at 120 minutes.

3.4 Influence of the exhaust gas to plant growth

The experiments were carried out twelve times, spending six month. The average height of all plants is 19cm and their statements were basically same and there was no branches had grown in all four schefflera plants before experiment. And four schefflera plants all grew well after the last experiment. The growth statements of all plants were shown in Fig. 7.

Fig. 7(a) and 7(b) directly showed that schefflera plants growth status in experimental group and control group after the sixth time experiment. The average height of two groups is still 19cm, which do not change apparently. It was concluded that exhaust gas of catalytic combustion does not have obvious effect on boosting plant growth at that time. There are four new branches in Fig. 7(c), twice as much as it shown in Fig. 7(d). Schefflera plant in experimental group grew more sprouts than that in control group, which means the exhaust gas of catalytic combustion could help new branches grow faster. After a-month experiment, the result showed difference of growth statement between two groups. Sequentially, the experiments were done for another six times during the period of five month. The average height of experimental group increased to 22 cm, higher by 3cm than that of con-

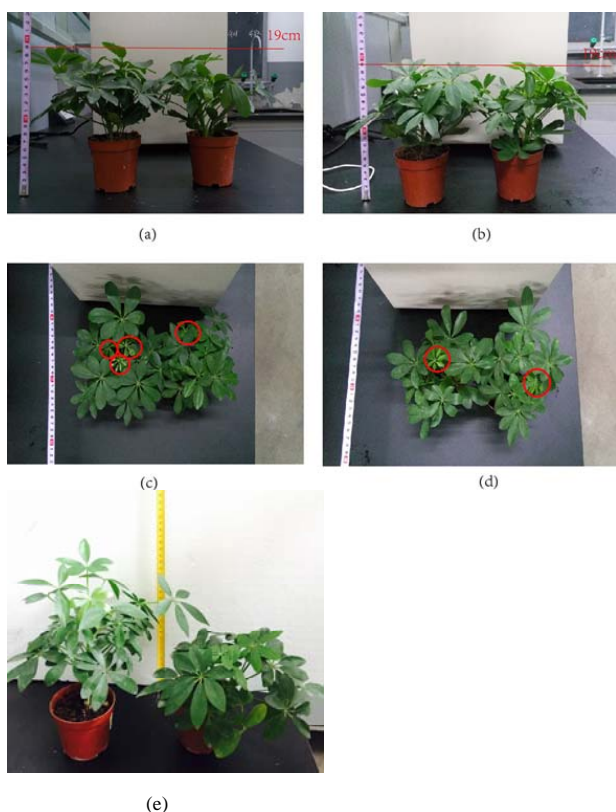


Fig. 7 (a)Schefflera plant growth status in experimental group after the sixth experiment, (b)Schefflera plant growth status in control group after the sixth experiment, (c)Sprouts in experimental group after the sixth experiment, (d)Sprouts in control group after the sixth experiment, and (e) Schefflera plant growth status of the experimental group(left)and the control group(right) after the last experiment.

control group shown in Fig. 7(e). The result showed that schefflera plant in experimental group grow taller and greener.

The temperature in greenhouse shown in Fig 8 at atmospheric pressure and at steady state was studied experimentally. The temperature change between 27.5 °C to 29.5 °C when environment temperature and humidity were 26.5 °C and 41 %RH respectively.

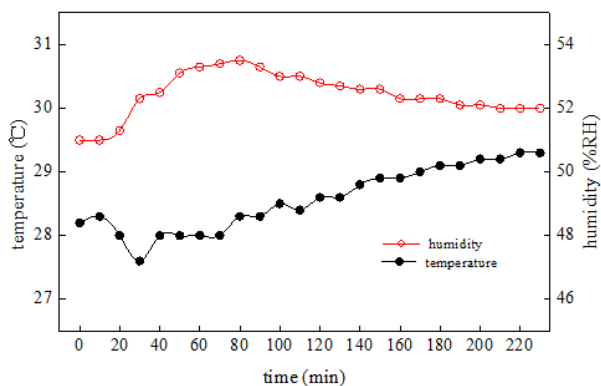


Fig. 8 Curves of temperature and humidity in greenhouse

The temperature in greenhouse rose slowly due to closing the furnace door after 70 minutes. During the whole experiment, the change of temperature is very small, which is close to room temperature. While the humidity in greenhouse shed varying from 50%RH to 54%RH also was showed in figure 8. The humidity began to rise slightly after

closing the furnace door at 70 minutes because there was a large amount of water vapor generation during combustion.

Figure 9 plots the concentration of CO₂ inside greenhouse. CO₂ increased before 70 minutes, then it changed between 3850 mg/m³ and 3950 mg/m³ after 70 minutes. Obviously, the CO₂ content of greenhouse was much higher than that of indoor air due to carbon dioxide produced by catalytic combustion.

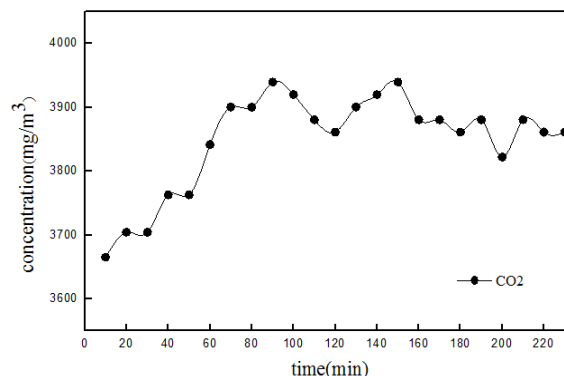


Fig. 9 Curves of the CO₂ content in the greenhouse shed

4. CONCLUSIONS

The radiation fluxes of catalytic burner were higher than that of conventional one because of high apparent area surface of catalytic monolith. The radiation efficiency of the monolith alone varied from approximately 20% to 40%. The radiation characteristics of the catalytic combustion furnace were analyzed by calculating radiation heat transfer in an enclosed furnace. It was found that the high radiation heat transfer in an enclosed furnace played an important role to heating glazed tiles. The glazed tile products heated by the catalytic combustion furnace are more smooth and glossy than that of conventional ones. At the same time its bisque was combined with glaze very well. Furthermore, it proved that the pollutant emissions (NO_x and CO) of exhaust gas were closed to zero while the catalytic combustion of furnace reached a steady state.

Besides, the influence of catalytic combustion exhaust gas to plant was also discussed by means of comparison. The schefflera plant in experimental group grew more sprouts and taller than that in control group. The exhaust gas of catalytic combustion with 6% carbon dioxide has a promotion effect on the growth of schefflera plants. The exhaust gas of catalytic combustion furnace could be applied to agriculture-greenhouses. It indicated the advantages of energy-saving and environmental protection for the catalytic combustion. Catalytic combustion played an increasingly important role for lower carbon emissions in industrial applications.

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NOMENCLATURE

A	apparent area of the monolith at the open end (m ²)
A_1	apparent area of the monolith at the open end (m ²)
A_2	apparent area of the wall surface (m ²)
$CV_{CH_4,net}$	net calorific value of CH ₄
F	view factor
\dot{V}_{CH_4}	volume flow rate of CH ₄ (N·m ³ /s)
$X_{1,2}$	shape factors

T_a	temperature of the surroundings (K)
T_s	average surface temperature (K)
T_1	average surface temperature of monoliths (K)
T_2	average temperature of the wall surface (K)

Greek Symbols

$\Phi_{1,2}$	radiation heat exchange (kW)
ϵ_1	emissivity of catalytic honeycomb monoliths surface
ϵ_2	emissivity of wall surface
ϵ_{eff}	effective emissivity
σ	the Stephan-Boltzmann constant

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