Research on energy-saving and exhaust gas emissions compared between catalytic combustion and gas-phase combustion of natural gas

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Abstract: In this paper, exhaust gas emissions were compared between conventional gas-phase combustion in both forced exhaust gas concentration of hot-water burner & premixed natural gas/air burner with heater and the catalytic combustion in catalytic honeycomb monolith burner. Test proved that the pollutant emissions of gas-phase combustion **were** higher than that of catalytic combustion. It is shown that the conversion of conventional gas-phase combustion **was** lower than that of catalytic combustion by measured experimental data. It indicated the advantages of energy-saving and environmental protection for the catalytic combustion.

Keywords: *catalytic combustion, exhaust gas analysis, near zero pollutant emissions, energy-saving*

1. Introduction

Catalytic combustion of natural gas has received considerable attention in the last decades due to its practical applications in both power generation and pollutant abatement[1-4]. This reaction has been shown to be effective in producing energy in gas turbine combustors. Compared to the conventional thermal combustion process, using a heterogeneous catalyst can remarkably decrease the reaction temperature, thereby reducing the noxious emissions of nitrogen oxides[5-6]. By enabling the combustion of extraordinarily lean fuel/air mixtures, the catalytic combustion of natural gas provides a low-emission alternative to gas-phase flames[7-9].

In this paper, exhaust gas emissions were compared between conventional gas-phase combustion and the catalytic combustion in catalytic honeycomb monolith burner VI. In order to study the exhaust gas of both forced exhaust gas concentration of hot-water burner & premixed natural gas/air burner with heater and catalytic combustion burner VI, their composition and content were measured, respectively. Meanwhile their combustion efficiency were calculated.

2. Experimental set-up and steps

Figure 1 illustrates the exhaust gas analysis system of catalytic combustion burner VI, The square honeycomb monoliths were 150mm in side of the square and 20mm long, with square-shaped cells which sectional area was 1mm×1mm. The support for all the monoliths tested here was cordierite. The four square catalytic honeycomb monoliths were installed in the burner VI each time. The lengths of catalytic honeycomb monoliths were 20mm for the catalytic combustion burner. 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.
In the experiment, 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 0~80m3/h of full-scale range , respectively. The two meters were provided electric current through manostat.

In the process of ignition, we need to swept the inside of burner VI by air for five minutes to ensure that there was no residual natural gas. In order to warm the honeycomb monoliths, the burner VI must be ignited by gas phase combustion with the excessive air coefficient at 1.3. When the catalytic surface came to be red, the excessive air coefficient should be adjusted to 2.0. Until it came into the steady state of catalytic combustion, the exhaust gas could be measured by the analyser. At the same time we observed and recorded the data.

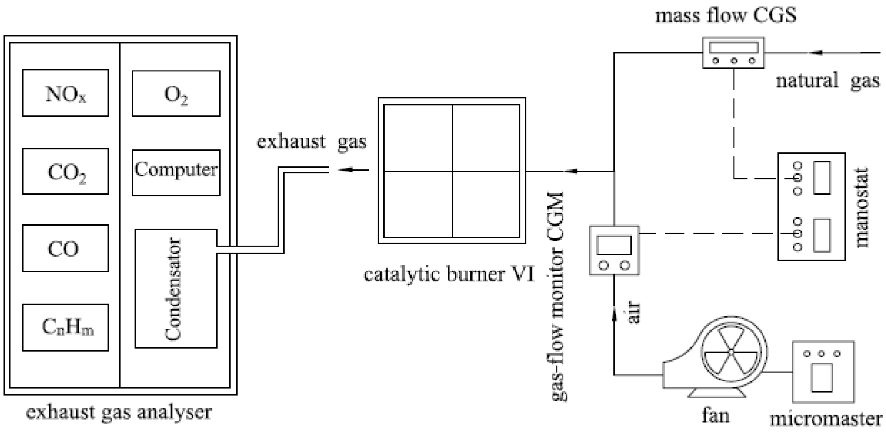

Fig. 1. Exhaust gas analysis system of catalytic combustion burner VI.

Figure 2(a) illustrates the exhaust gas analysis system of premixed natural gas/air burner with heater. This burner was ignited by gas phase combustion with the excessive air coefficient at 1.1 and 1.3. When the water heater came into the steady state of gas phase combustion, we observed and recorded the experiment data from its chimney. Also the Figure 2(b) illustrates forced exhaust gas concentration of hot-water burner with the excessive air coefficient at 1.3. and the exhaust gas concentrations were measured above the burner.

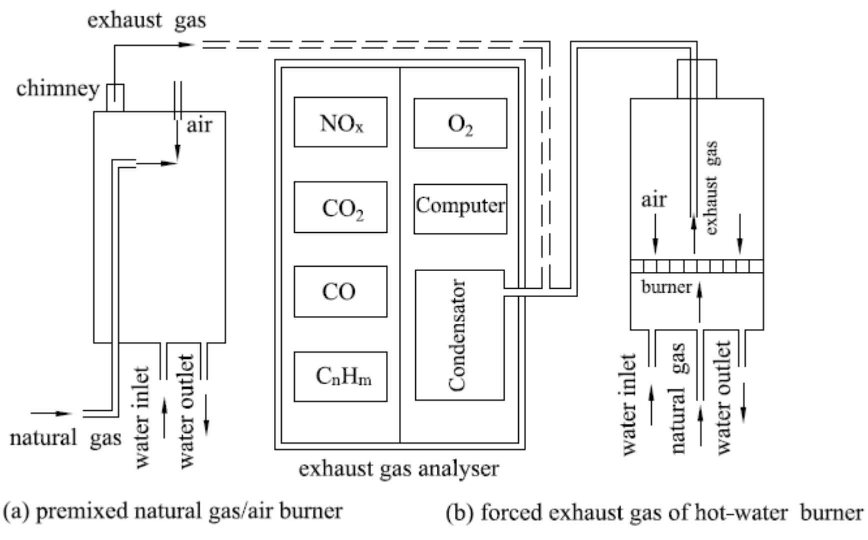

Fig. 2. Exhaust gas analysis system of premixed natural gas/air burner with heater and forced exhaust gas concentration of hot-water burner .

3. Results and Discussion

3.1. Emission characteristics of catalytic combustion and conventional gas-phase combustion

Form figure 3(a) plots the content of NOx was very low , because the temperature(T around 1000℃) of catalytic combustion and gas-phase combustion did not reach the degree which could generate a large number of heat-type NOx. But the emission of NOx in gas-phase combustion ascended gradually with the increase of natural gas flow rate.

The content of CO in catalytic combustion was very small **(closed to 0)**. From the data of CO **[figure 3(b)]** we got that the combustion efficiency of catalytic combustion burner VI is very high and its heat had been released fully. However, the content of CO of gas-phase combustion was higher than that of catalytic combustion. The maximum of CO emission reached about 150 ppm. It was shown that natural gas of gas-phase combustion did not oxidized completely.

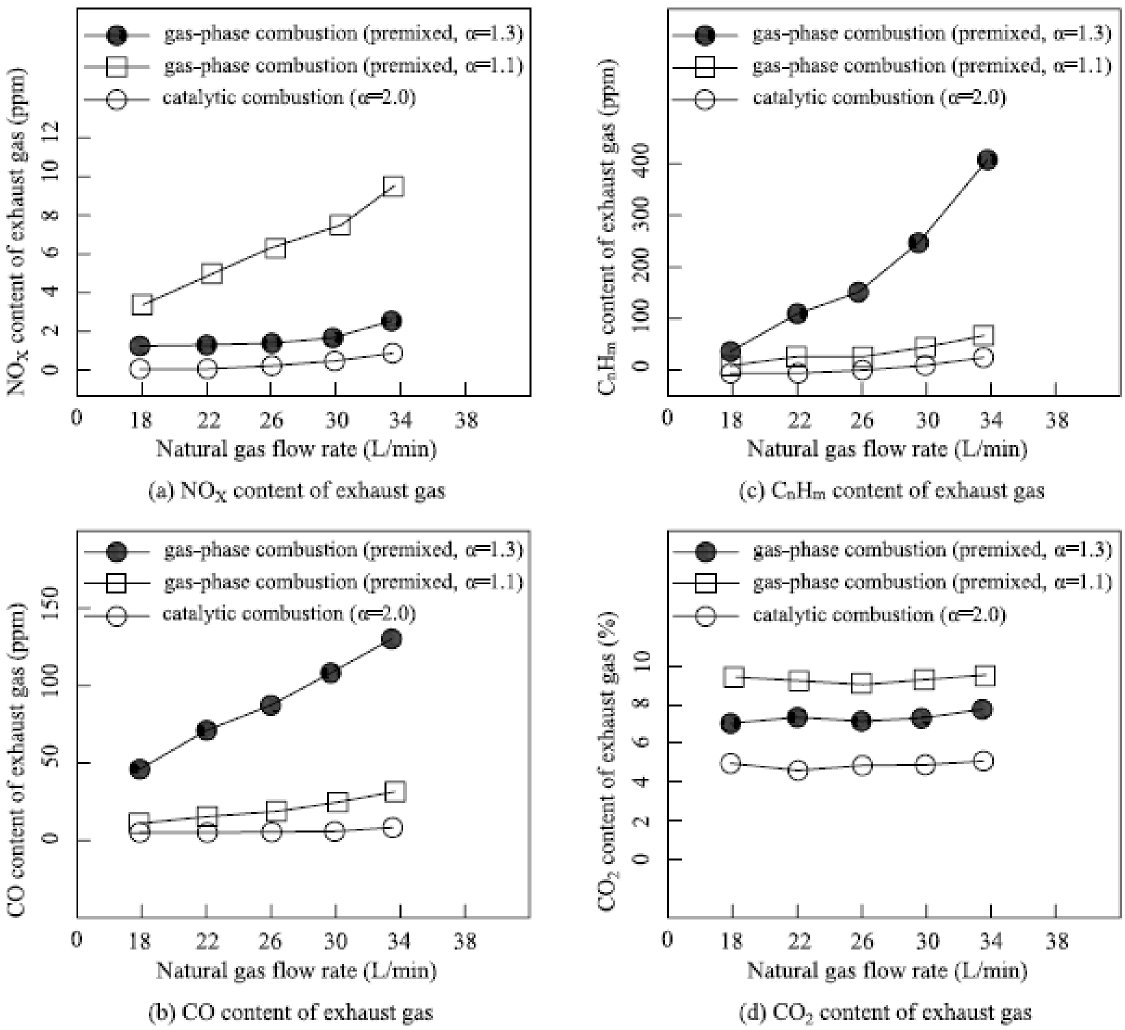

Fig. 3. The exhaust gas content of catalytic combustion and conventional gas-phase combustion （premixed, α=1.3 and 1.1） under the condition of different natural gas flow rate (α is the excessive air coefficient).

The figure 3(c) also shows that there was no C_nH_m from the exhaust gas of catalytic combustion. It was evidenced that the catalytic combustion efficiency was almost closed to 100%. It was seen that the content of C_nH_m in gas-phase combustion was 39.3 ppm～428 ppm. At the same time, C_nH_m was increased quickly with the increasing natural gas flow rate. It proved that the conversion of gas-phase combustion was lower than that of catalytic combustion.

When the excessive air coefficient decreased from 1.3 to 1.1 in premixed natural gas/air burner with heater the conversion of gas-phase combustion increased dramatically, saturating at near 100%, the CO decreased with very lower value (about 8 ppm) in the cases of the high temperature chamber of furnace under certain conditions. But the exhaust gas temperature and heat loss increased from its chimney **with the same water flow rate.** The emission of NOx was increased quickly with the increase of natural gas flow rate. Simultaneously, the nosie happened during the combustion of premixed natural gas/air burner and the blue flame changed gradually into dark red **one.**

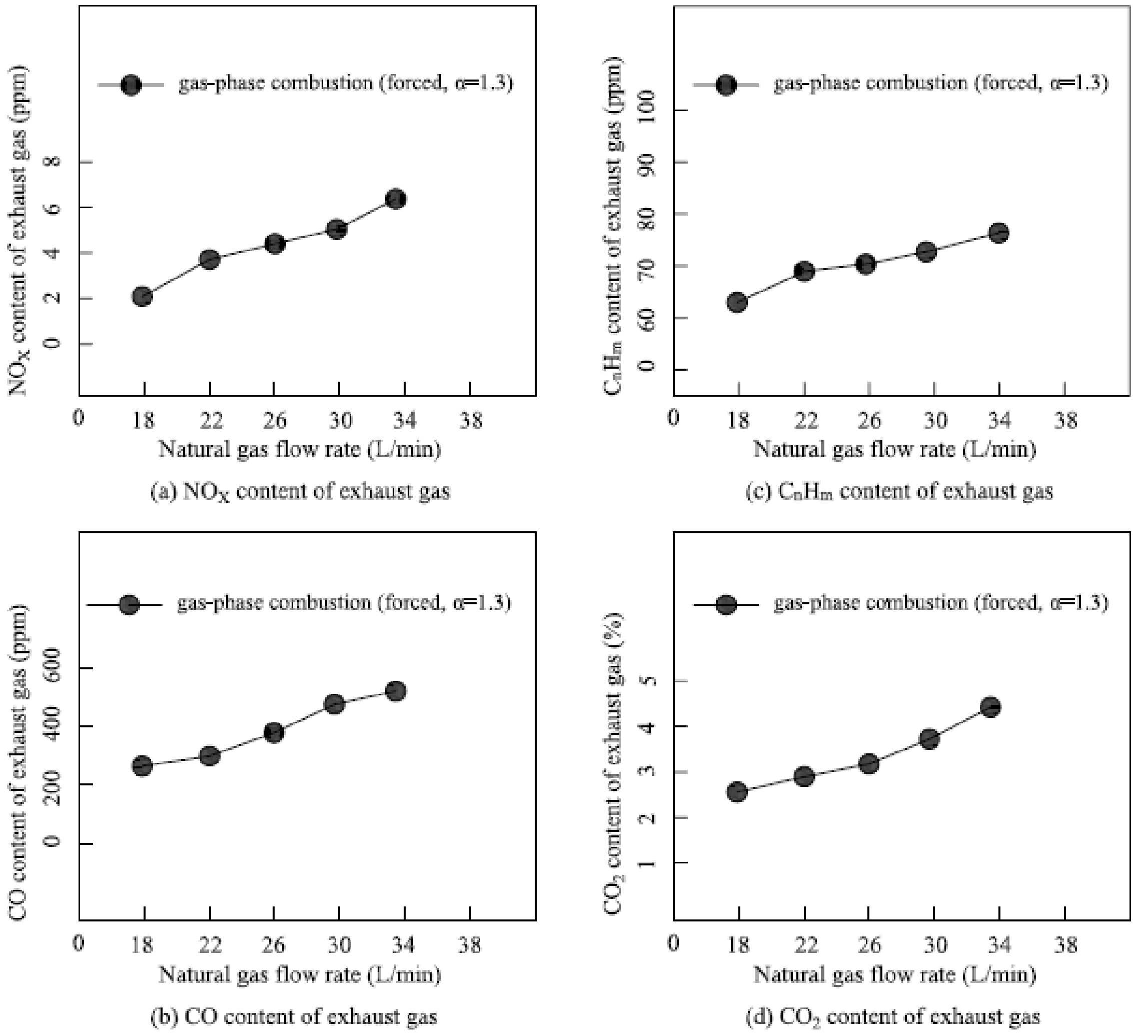

Fig. 4. The exhaust gas content of conventional gas-phase combustion（forced, a=1.3）under the condition of different natural gas flow rate.

The NOx、CO and un-burnt CH_4 concentrations in forced exhaust gas concentration of hot-water burner existed more with the excessive air coefficient 1.3. Its exhaust gas concentrations have been significantly diluted in large space by measured CO_2 data as shown in figure 4. Otherwise, the percentage of CO_2 should remain about 7-8% without vapor by CO_2 analyser.

For all tested of the catalytic combustion, only extremely small amount of CO, unburned fuel and NOx were detected inside the monolith channels and over the open

end of the burner VI. A catalytic combustion process can achieve 'near-zero' pollutant emissions.

3.2. Calculation of combustion efficiency in gas-phase combustion

It was evidenced that the catalytic combustion efficiency was almost closed to 100%. But there were a lot of C_nH_m and CO from the exhaust gas in gas-phase combustion. It proved that gas-phase combustion had not oxidized completely and its combustion efficiency should be calculated. As the main composition of natural gas was methane, so the chemical reaction equation is (1) in the following:

$$CH_4 + 2O_2 = CO_2 + 2H_2O \qquad (1)$$

CO was a kind of intermediate which was generated during combustion of hydrocarbons. The number of C atom remained unchanged in the reaction process. The total volume of CO and CO_2 were the same as that of CH_4 via the reaction equations (1) ($V_{CO} + V_{CO_2} = V_{CH_4}$). Given the volume of methane was 1 Nm^3 (V_{CH4}=1). According to equations (2), the volume of CO was calculated as:

$$\begin{cases} V_{CO} + V_{CO_2} = 1Nm^3 \\ \dfrac{V_{CO}}{V_f^d} = \gamma_{CO} \\ \dfrac{V_{CO_2}}{V_f^d} = \gamma_{CO_2} \\ \dfrac{V_{CH_4}^1}{V_f^d} = \gamma_{CH_4}^1 \end{cases} \qquad (2)$$

Where V_{CO} and V_{CO_2} are the volume of CO and CO_2 in exhaust gas(m^3), respectively. V_f^d is the total volume of exhaust gas. γ_{CO} and γ_{CO2} are ratio of CO volume and CO_2 volume to that of exhaust gas, respectively. $V_{CH_4}^1$ is the volume of unburnt CH_4 in exhaust gas(m^3). $\gamma_{CH_4}^1$ is ratio of CH_4 volume to that of exhaust gas. $\gamma_{CO}, \gamma_{CO2}, \gamma_{CH_4}^1$ are measured by the analyser.

According to equation (3): For $1m^3$ CO oxidized completely to CO_2 could generate 12644 kJ heat (H_2=12644 kJ). It proved that the content of CO had an important influence for utilization of thermal energy of the fuel.

$$\mathrm{CO} + 0.5\mathrm{O}_2 = \mathrm{CO}_2 + \Delta T \qquad (3)$$

So, the heat released of unburnt CH_4 and CO were calculated as:

$$Q_{CH_4} = V_{CH_4}^1 \times H_1 \qquad (4)$$

$$Q_{CO} = V_{CO} \times H_2 \qquad (5)$$

Where H_1 is net calorific value of methane under standard conditions which is 33.70MJ/Nm^3. H_2 is calorific value of CO which is 12644kJ/m^3.

The following equations(6) for heat released percent of unburnt CH_4 and CO to reactant (natural gas) was derived:

$$K = \frac{Q_{CH_4} + Q_{CO}}{V_{CH_4} \times H_1} \qquad (6)$$

The combustion efficiency of gas-phase was calculated by equation (7):

$$\eta = 1 - \frac{V_{CH_4}^1 + V_{CH_4}^2}{V_{CH_4}} \qquad (7)$$

Where $V_{CH_4}^2$ is the volume of CH_4 which has been used in generating CO, which was $V_{CH_4}^2 = V_{CO}$.

According to above equations and experimental data, table 1 shows ratio of heat released of unburnt CH_4 and CO and combustion efficiency under the condition of different natural gas flow rate. It was seen that part of the energy were wasted in gas combustion which did not oxidized completely.

Table 1. Ratio of heat released of unburnt CH_4 and CO and combustion efficiency

Natural gas flow rate		L/min	18	22	26	30	34
forced, a=1.3	k_1	%	0.58	0.63	0.65	0.66	0.67
	η_1	%	98.82	98.7	98.63	98.55	98.51
premixed, a=1.3	k_2	%	0.078	0.17	0.27	0.41	0.62
	η_2	%	99.88	99.76	99.66	99.49	99.27
premixed, a=1.1	k_3	10^{-2} %	0.94	1.00	1.05	1.10	1.20
	η_3	%	99.987	99.986	99.984	99.983	99.981

4. Conclusions

Exhaust gas emissions were compared between gas-phase combustion and catalytic combustion in catalytic honeycomb monolith burner VI. It proved that the concentration of pollutant emissions of gas-phase combustion were morehigher than that of catalytic combustion. It was shown that the conversion of gas-phase combustion was lower than that of catalytic combustion by calculated data. It can be concluded that catalytic combustion was completed oxidation combustion of the heterogeneous reaction. The emissions of NOx, unburnt C_nH_m, CO was very small, so the catalytic combustion would not cause serious environmental pollution. Therefore, High combustion efficiency and near zero pollution emissions of catalytic honeycomb monolith burner VI were its advantages which should be applied for industry.

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