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Development of a Catalytic Combustor with Heat Exchanger

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Abstract−Catalytic combustion is thought to be a considerable improvement on the traditional one under specific conditions. Due to its special features, catalytic combustion has two strong points compared to flame: no NO_x emission and high reaction efficiency. However, the preheating process of catalytic combustion is an obstacle that deteriorates profitability in operation. So the HTHE (High Temperature Heat Exchanger) is adapted to the system to reinforce the preheating process, and we show that the catalytic combustion is maintained steadily without exceptional heat injection. As a result, the stability on the catalytic surface is the most important operational factor. To achieve it, both mixture gas property and temperature distribution should be controlled.

Key words: Catalytic Heat Exchanger, Catalytic Combustion, Fin Tube, Catalyst

INTRODUCTION

A catalytic combustor with a heat exchanger has been employed in various manufacturing processes that require a compact design [Klein and Eigenberger, 2001]. Fin tubes have been widely used to augment the surface area, which is a determinant factor for increasing the heat transfer rate. Wang [2000] extensively reviewed the recent progress and performance of the fin-tube heat exchanger.

Wang et al. [2001] investigated catalytic combustion involving a strongly exothermic reaction such as LNG combustion in a tubular reactor with a fixed catalyst bed inside the tube and a cooling fluid outside. They found that a large number of tubes caused a severe pressure drop due to catalyst bed, non-uniform distribution of heat transfer across the radial direction, a multiplicity of hydrodynamic regimes, etc. Seo et al. [2000] designed a catalytic heat exchanger using catalytic fin tubes catalyzed with Pd catalyst and found that the arrangement of the fin tubes had a significant influence in achieving best catalytic combustion performance. When the heat exchanger was enlarged to a size corresponding to 100,000 kcal/hr, the catalytic fin-tubes showed unsatisfactory performance in catalytic combustion and heat exchange rate. Thus, they replaced the catalytic fin tubes with catalyzed honeycombs to increase the heat exchange capacity.

A catalytic combustor with a catalyzed honeycomb can be effectively combined with a heat exchanger. The catalytic combustion of the mixture of air-fuel also can be improved by employing a regenerative preheating system. Catalytic combustion also has the unique feature of stably burning a very lean mixture that cannot be combusted with conventional flame combustion. Thus, the appropriate use of catalytic combustion would control the combustion temperature below 1,000 °C, which prevents the production of thermal NO_x [Seo et al., 1999; Schlegel et al., 1996]. Catalytic combustion as an environmentally friendly technology has been applied to various facilities such as gas turbines, industrial boilers, radiant heaters, VOC abatement etc. [Trimm, 1995; Saint-Just and Kinderen, 1997; Sadamori, 1996].

In the present work, a catalytic heat exchanger was designed which employs the regenerative preheating system of combustion air. The catalytic heat exchanger is composed of a catalytic combustor and a heat exchanger. The sensitivity of operating parameters such as equivalence ratio, velocity and preheating temperature of the mixture of air-fuel was investigated to analyze the combustion characteristics in the catalytic combustor. One of the objectives in this work is to find optimal operating conditions for stable catalytic combustion in the catalytic heat exchanger.

EXPERIMENTAL

The regenerative heat exchanger is composed of two parts: a catalytic combustor and a heat exchanger. The catalytic combustor consists of a preheating burner and catalyzed honeycombs. The heat exchanger is composed of one for the preheating of the mixture and the other for working fluid. Fig. 1 shows the concept of the catalytic heat exchanger used in this study. The system was designed

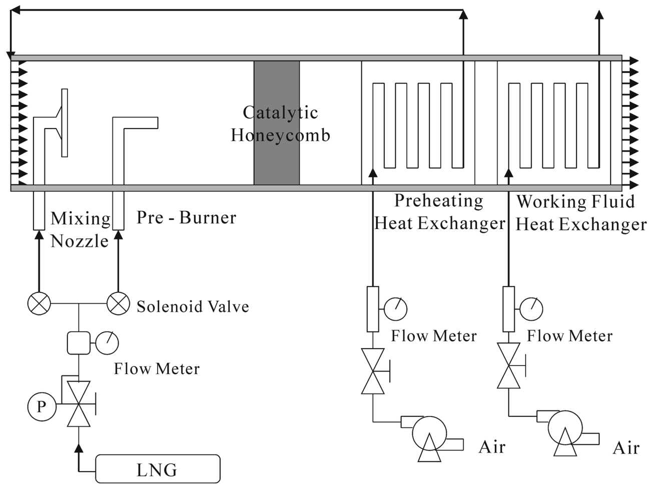

Fig. 1. Schematic diagram of the test rig for catalytic heat exchanger.

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to utilize the heat generated by catalytic combustion as a heating source of the mixture fed to the catalytic combustor and to recover the remaining heat via a heat exchanger. To initiate the catalytic combustion upon the startup of operation, a pre-burner was installed at the place before the catalytic combustor. It is only used upon the startup of the system and turned off when the catalytic combustion is ignited completely.

The casing of the heat exchanger was made of SUS 304 to withstand high temperatures up to 1,000 °C. Ceramic fiber was used to insulate the system. The fin tubes of the heat exchanger were fabricated to the triangular-staggered type. The heat exchangers for the preheating of the mixture and the heat recovery have a surface area of 20 m^2 and 40 m^2, respectively. To prepare the catalytic honeycombs, the ceramic honeycomb (150 cells/in^2) was first washcoated with TiO_2 of high surface area. Then Pd catalysts were deposited to the washcoated honeycombs by impregnation with aqueous palladium nitrate ($Pd(NO_3)_2$) (19.96% Pd, Engelhard). The catalytic honeycombs were dried at 100 °C for 12 h and calcined at 550 °C for 6 h. The catalyst loading on the catalytic honeycombs was about 1.0 wt%.

LNG (CH_4 90.22%, C_2H_6 6.45%, C_3H_8 2.34%, C_4H_{10} 0.99%) was used as a fuel and its flow rate was measured with a dry typed flowmeter. The LNG was supplied to the preheat burner and the mixture nozzle. Air was supplied to two places-the inlet of the system and the heat exchanger recovering the generated heat- by turbo-blowers of which each flowrate is 600 Nm^3/hr. A diffusion type burner was used as a preheating burner. The fuel feed to the preheating burner was controlled by remote control depending on the operating situation of the catalytic heat exchanger. The mixing nozzle was designed to have the mixture of LNG-air mixed as homogeneously as possible because the mixing degree of the mixture has a critical effect on the catalytic combustion. The catalytic combustor was fabricated with catalytic honeycombs and installed downstream of the pre-burner. A sight glass was installed just behind the catalytic combustor to observe the situation of the catalytic honeycomb during the operation of the system.

The experiment was conducted according to the following procedure. The combustion air is fed to the inlet of the system through the preheating heat exchanger. LNG is supplied to the pre-burner whereas the mixing nozzle is closed. The pre-burner is ignited so that the combustion air is heated to a level that can initiate the catalytic reaction in the catalytic combustor. When the temperature of the combustion air reaches the ignition level, 350 °C, LNG is supplied to the mixing nozzle and is mixed with the on-going combustion air. In the catalytic combustor, the mixture is heated by the catalytic reaction onto the catalytic surface. The combustion gas exiting from the catalytic combustor flows to the preheating heat exchanger, where the heat is transferred from the combustion gas to the combustion air flowing inside the fin tube. The combustion gas flows further to the downstream, where another heat exchanger is placed. The remaining heat in the combustion gas is recovered via this heat exchanger of which the working fluid is the air and the combustion gas is exhausted to the atmosphere.

The temperatures of the combustion gas and the surface of the

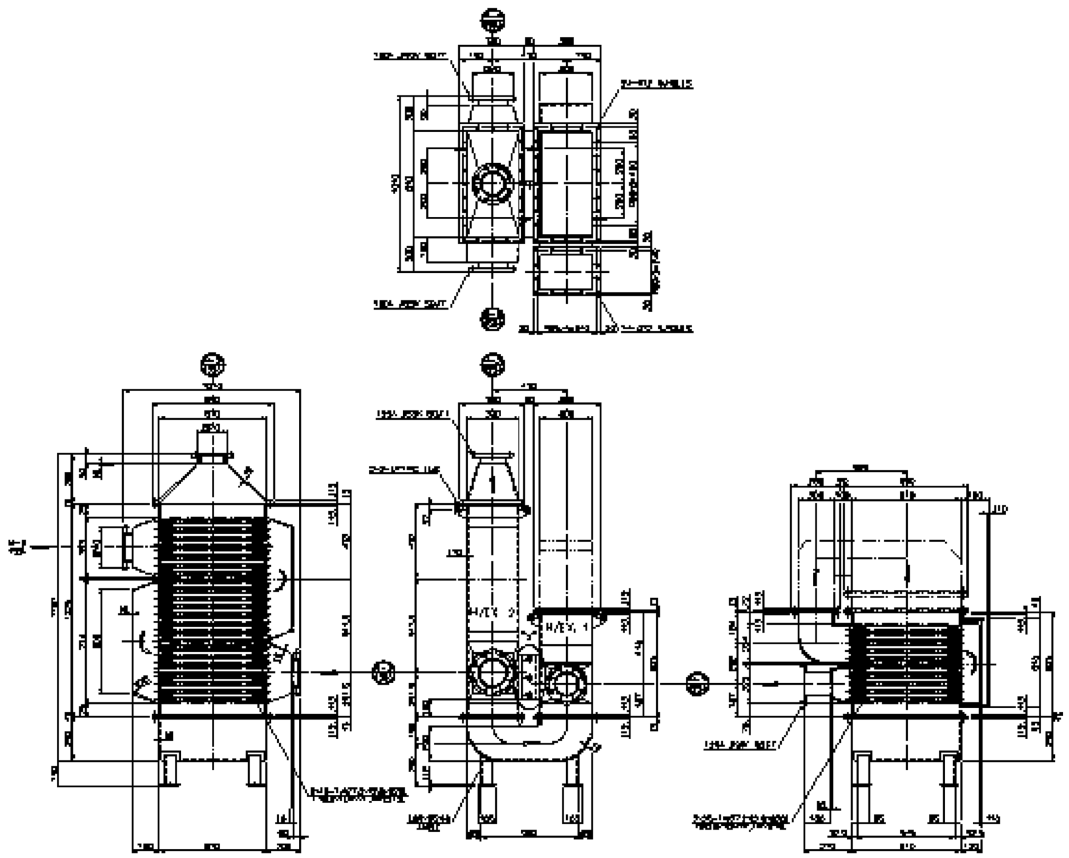

Fig. 2. Design of the regenerative catalytic heat exchanger.

catalyst honeycomb were measured with K-type thermocouples. The combustion gas was sampled at the exit of the system and analyzed with an on-line gas analyzer. The concentration of CO_2 and CO was measured with the NDIR analyzer (Horiba, VIA 510). THC concentration was measured with FID analyzer (Horiba, FIA 510). Water in sampling gases was removed by a sample conditioner before the analysis was performed.

RESULTS AND DISCUSSION

1. Effects of the Preheating Heating Temperature

The temperatures within the catalytic heat exchanger and the compositions of the combustion gas were measured. The conversion of combustion gas was calculated by the following equation:

$$\text{Conversion (\%)} = \frac{[CO_2]}{[HC]+[CO_2]} \times 100 \qquad (1)$$

The preheating temperature of the mixture was changed in the range of 300-420 °C at a flow rate of 408.2 m^3/h and an equivalence ratio of 0.19-0.27 (see Fig. 3). The preheating temperatures of the mixture were measured at the upstream of the catalytic honeycomb. The results show that the preheating temperature significantly affects the conversion of the mixture. The conversion rate, in general, increases with the increase of the preheating temperature. Especially, the conversion rate is abruptly changed at a preheating temperature of 380 °C for all ranges of equivalence ratio. It reaches over 98% when the preheating temperature increases to over 380 °C. The results suggest that the preheating temperature should be kept over 380 °C to obtain nearly complete conversion of the mixture.

2. Effects of the Mixture Velocity

Seo et al. [2000] investigated the catalytic combustor using catalytic fin tubes and found that the mixture velocity was a critical factor in the design of the catalytic heat exchanger. The conversion rate seems to be very sensitive to the contact time of the mixture on the catalyst surface. The effects of the mixture velocity on the conversion rate were investigated as shown in Fig. 4. The mixture velocity was changed in the range of 0.52 to 0.77 m/s at a preheating temperature of 380 °C and an equivalence ratio of 0.19-0.27. As a result, the conversion rate appears to be over 98% in all the experimental conditions. Specifically the conversion rate is a little decreased from 99% to 98% when the mixture velocity is raised from 0.52 m/s to 0.77 m/s. These results verify that the mixture velocity of 0.52-0.77 m/s is the appropriate operating condition for the manufactured catalytic combustor.

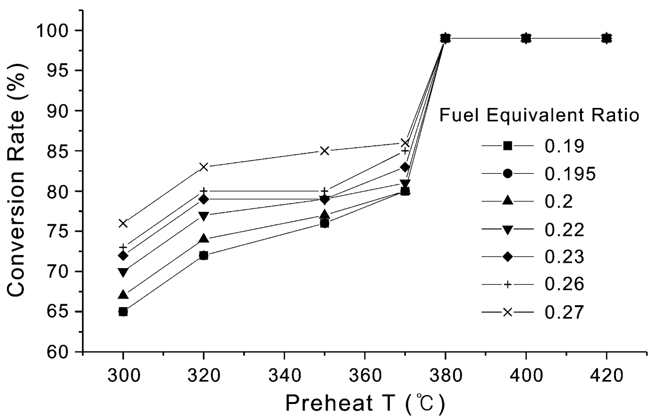

Fig. 3. The effects of the preheating temperature of the mixture gas on the conversion rate.

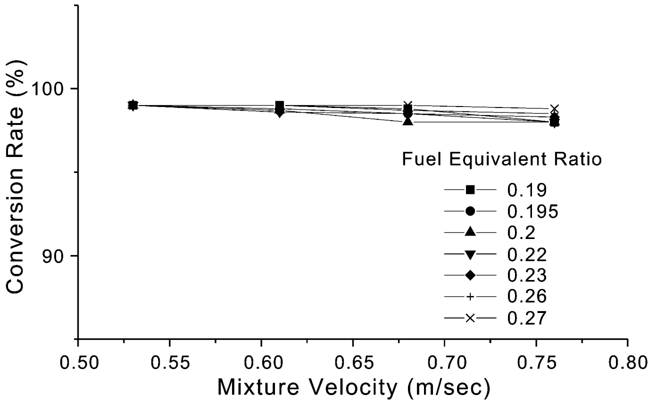

Fig. 4. The effects of the mixture velocity on the conversion rate.

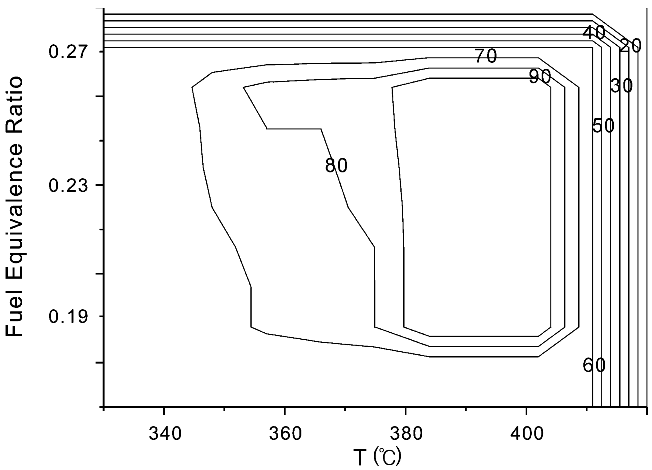

Fig. 5. The contour diagram of the conversion of the mixture as a function of equivalence ratio and preheating temperature. The number in the contour refers to the conversion rate of the mixture.

3. Effects of the Equivalence Ratio of the Mixture

The influence of the equivalence ratio on the conversion was investigated in the range of an equivalence ratio of 0.16-0.3 at a flow rate of 408.2 m^3/h. Fig. 5 represents the conversion rate as a function of both the equivalence ratio and the preheating temperature. In the flame combustion of CH_4, the lean flammability limit is an equivalence ratio of 0.5. But the results show that a catalytic reaction is possible even at a very low equivalence ratio of 0.2, which is very lean compared to the lean limit of the conventional flame combustion.

As a result of the experiment, the conversion rate reaches over 98% at a high preheating temperature of over 380 °C and an equivalence ratio of 0.19-0.27. But, unstable operation was observed when the equivalence ratio exceeded the range of 0.19-0.27. The most stable catalytic combustion was achieved at an equivalence ratio of around 0.25. This implies that stable catalytic combustion is possi-

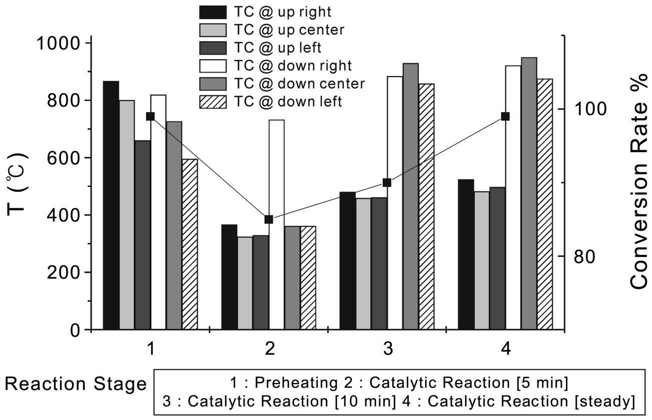

Fig. 6. The temperature distribution in the catalytic honeycomb layer and the conversion rate of the mixture at each operational sequential process.

ble within a limited range of equivalence ratio owing to the heat balance in the full system. Both overheating and the under-heating can be troubles encountered in the design of the catalytic heat exchanger.

4. Temperature Distribution of the Catalytic Honeycomb Surface

The temperature distributions on the catalytic honeycomb surface were different depending on each ignition step of the catalytic combustor. Fig. 6 shows the change of the temperature distributions at the inlet and outlet surface of the catalytic combustor, and the conversion rate according to the ignition process of the catalytic combustor. For the preheating step, the temperature at the inlet surface of the catalytic combustor appeared higher than that of the outlet surface. However, when the catalytic combustion in the catalytic combustor reached a stable condition, the temperature at the outlet surface of the catalytic honeycomb was relatively higher than that of the inlet surface.

In the step to preheat the catalytic combustor, the averaged temperature at the inlet and outlet surfaces of the catalytic honeycomb was relatively higher compared to other operation steps. After some time of preheating, catalytic combustion is initiated and the temperature distributions at both the inlet and outlet layer of the catalytic honeycomb become more uniform. The conversion rate is steadily increased while the catalytic combustion in the catalytic combustor is becoming stable. When the catalytic combustion becomes completely stable, the conversion rate reaches about 99%. At that time, the peak temperature in the catalytic honeycomb is 50 °C higher than in the preheating step. The results imply that the conversion rate becomes higher as the peak temperature of the honeycomb layer gets higher.

CONCLUSIONS

A catalytic heat exchanger was designed that employs the regenerative preheating system of combustion air. The results show that the mixture velocity does not significantly affect the performance of the catalytic combustor, whereas the preheating temperature of combustion air significantly affected the conversion rate. Complete conversion was achieved in the catalyzed honeycomb at a preheating temperature of 370-390 °C, a mixture velocity of 0.53-0.75 m/s and an equivalence ratio of 0.19-0.27. The heat exchange efficiency of the catalytic heat exchanger appeared to be about 75% when the air at room temperature was used as a working fluid. The results show that both the temperature distribution on the catalytic surface and the mixture conditions determine its stable catalytic combustion.

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