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The Development of Catalytic Combustor With Heat Exchanger

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1. Introduction

Catalytic combustor with heat exchangers are often employed in process technology where a compact design is required [1]. The use of fin and tube heat exchanger offers the enhanced surface area for heat exchange. The recent progress and performance of the fin-tube heat exchanger, especially airside, has been reviewed extensively by Wang [2].

Catalytic processes involving strongly exothermic reactions such as LNG combustion, 10,000 kcal/Nm3, are performed in tubular reactors with a fixed catalyst bed inside the tube and a cooling agent flowing outside. Many problems for the use of a large number of tubes are a pressure drop due to catalyst bed, heterogeneous distribution of heat transfer along the radial direction, a strong sensitivity of the process and a multiplicity of hydrodynamic regimes, etc. [3].

The coating of active catalyst onto the fin side resulting in uniform close contact of heat release and heat exchange surface has been found to eliminate the problem of the pressure drop, and heat transfer. Then, the thermal conductivity of fin-tube and design of multiple fin-tubes heat exchanger appears important for the device application [4].

However, for scale up of the device, above 100,000 kcal/hr, the active catalyst coating on fin-tube was found to show an unsatisfactory performance of heat exchange and conversion efficiency of fuel. Increasing the heat exchange capacity is necessary to enlarge the amount of catalyst, resulting in the use of catalyzed honeycomb instead of active catalyst coating onto the fin-tube. The catalyzed honeycomb can be combined compactly with multiple fin-tube heat exchanger. The conversion efficiency of LNG can also be improved employing a regenerative air preheating system.

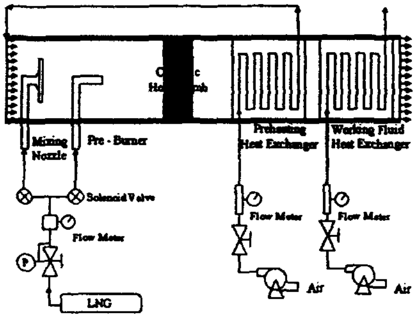

Fig.1 Schematic Diagram for Catalytic Heat Exchanger

The catalytic combustion of LNG lowers the combustion temperature, ~ 1000 C, compared to that of flame combustion, 1500 C, avoiding generation of NOx below few ppm level and making possible to use conventional SUS304 steel as a heat exchanger and device parts [5]. This catalytic combustion as a new emerging clean and environmentally friendly technology has been applied to gas turbine, industrial boiler, VOC abatements etc. [6-11].

In present work, the analysis of the combustion characteristics in the catalytic heat exchanger has been performed to investigate the sensitivity of flow control such as mixture composition, velocity and preheating of air. Throughout the work, the LNG as fuel and air as working fluid used to test the feasibility of the system configuration employing multiple fin-tubes for heat exchange and a regenerative preheating part for air shown in Fig. 1. We aims in this work to find a optimum stable catalytic combustion range for the performance of catalytic heat exchanger.

2. Experimental

The experimental equipment is mainly composed of two sub-parts: the combustion part and the heat exchange part. The combustion part is composed of the preheating burner part and the catalytic combustor part. And the heat exchange part is composed of the mixture preheating part and the working fluid heating part. The figure.1 describes the catalytic heat exchanger which was designed and manufactured to perform the experiments. This equipment tried to utilize the heat generated by catalytic combustion for mixture preheating and heat transfer to the working fluid simultaneously. To achieve these goals it was designed to use the preheating burner to heat the mixture gas up at only initial stage.

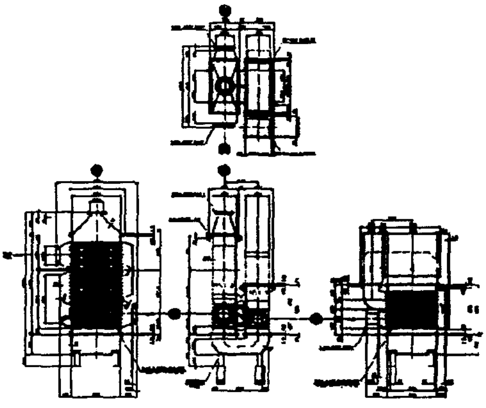

Fig. 2 Design of Catalytic Combustor with Heat Exchanger

The main body of the equipment is made of SUS 304 steel to stand the high temperature up to 1000oC partly. And the ceramic thermal barrier was used to avoid losing the heat. Heat exchange fin tube arrayed in the form of a staggered type at the heat exchange part. The heat exchange fin tube is made of SUS 304 steel and has each area of 20 m2 , 40 m2 . Flow system is shown in Fig. 1. To prepare for the catalytic honeycombs, the ceramic honeycomb (150cells/inch2) were first washcoated with TiO_2 of high surface area. Then Pd catalysts were added to the washcoated honeycombs by impregnation with aqueous palladium nitrate ($Pd(NO_3)_2$) (19.96% Pd, Engelhard). The catalytic fin tubes were dried at 100 oC for 12 h and calcined at 550 oC for 6 h. The catalyst loading on the catalytic honeycombs was about 1.0 wt.%.

The LNG (CH_4 90.22%, C_2H_6 6.45%, C_3H_8 2.34%, C_4H_{10} 0.99%) used as a fuel was supplied from reservoir and it was measured with an accumulating type flowmeter. The LNG flew through a solenoid valve which was controlled in according to the experimental process. The LNG was supplied to whether the preheat burner or the mixture nozzle in according to the experimental process. The air which was used as a mixture and a working fluid was supplied by two fan type blowers of which each capacity was maximum 600Nm3/hr. Diffusion type burner which was composed of an igniter and a fuel injection pipe was used as the preheating burner. The fuel injection onto the burner was controlled by a control panel electrically. The mixer nozzle was used to make the mixture homogeneous because the mixing degree of the mixture has an crucial effect on the combustion characteristics. The nozzle was designed considering the ejection cross area in according to the gradient of pressure. The catalytic combustor was installed with the honeycomb deposited with Pd catalyst (1wt%). The honeycomb with 150 cells per square inches maximized the surface area against mixture. Moreover, with avoiding the interruption of mixture flow, the watching window was installed to observe the combustion phenomena directly.

The experiment was developed in according to this process. The airflow produced by the blower arrived at the preheating heat exchanger through a valve and a flow meter. The airflow passing through preheating heat exchanger was heated by flame generated by preheat burner. The heated air passed through the catalytic layer and then arrived at preheating heat exchange fin tube surface. Then, the heated air and combustion gas generated by preheating burner transferred the heat to the preheating heat exchange fin tube. After then, these gases were emitted out through the surface of the working fluid heat exchange fin tube bank.

As the process was going on like above continuously, the temperature of the preheating air passed through the preheating heat exchanger was getting to be warm up to the catalytic reaction starting temperature 350oC (LNG based). When the temperature of preheating air got to the catalytic reaction starting temperature, the fuel supply to the preheating burner was quitted and then the fuel supply to the mixing nozzle situated in front of the catalytic combustor started. The mixture composed of the air and fuel which are mixed by mixing nozzle started to react on the catalytic surface layer. Being discharged out , the exhaust gas which was generated by the catalytic reaction accomplished the heat transfer into the heat exchange part.

At each point which had important meanings experimentally, the temperature of the gas and the surface of the catalyst was measured with K-type thermocouples. The exhaust gas was collected and investigated at the exhausting exit. The concentration of the CO2 and CO was investigated with the NDIR analyzer (HORIBA , VIA 510). And THC concentration was measured with FID analyzer (HORIBA, FIA 510).

3. Results and Discussion

The temperature distribution within the catalytic heat exchanger and the composition of combustion gas were measured. The conversion of combustion gas which was introduced to investigate the combustion characteristics was calculated by the following equation (1)

$$ConversionRate(\%) = \frac{CO2}{HC + CO2} \quad ...(1)$$

Each measured value is the mean value in the steady state.

3.1 The combustion characteristics on the mixture gas preheating.

The preheating temperature of the mixture gas was changed from 300°C to 420°C in the condition of mixture gas flow rate 408.2 m3/h and the fuel equivalence ratio 0.19 ~ 0.27 .The preheating temperature of mixture gas was measured as the basis of the point in front of the preheating burner. This was for taking the measured temperature of the mixture gas off distortion by the flame of the preheating burner .

The conversion rate had the value over 98% when the mixture preheating temperature was over 380°C. When the mixture preheating temperature was around 350 °C, the combustion reaction began, but the catalytic combustion wasn't able to be maintained steadily with showing relatively low conversion rate. This means that at the stage that the flame converted to the catalytic reaction the compensation of some amount of the heat in the mixture gas is required in order to achieve the steady state reaction. This was due to the heat loss caused by the gap between converting heat source.

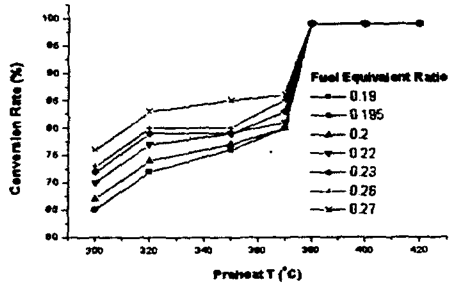

Fig. 3 Thc combustion characteristics on the preheating temperature of the mixture gas

3.2 The combustion characteristics on the mixture gas velocity

According to the fin tube type catalytic combustor experiments[4] the velocity of the mixture gas is the crucial factor which determine the capacity of application. This is caused by the reason that the conversion rate is very sensitive to the staying time of the mixture on the catalyst surface as the surface reaction. The velocity of the mixture gas was changed in the condition of the preheating temperature 380°C , the fuel equivalence ration 0.19 ~ 0.27 . As a result, there was little change in the conversion rate due to the sufficient catalyst surface area to the given conditions. Therefore honeycomb type catalytic supporter is better than the fin tube type catalytic supporter in securing the reaction area.

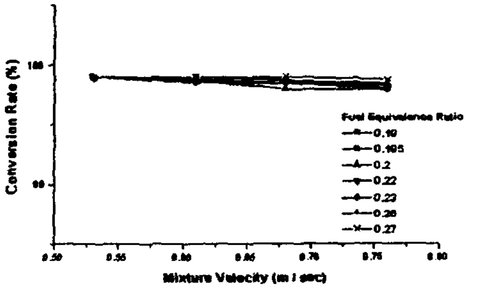

FIg. 4 The combustion characteristics on the velocity of the mixture gas.

3.3 The combustion characteristics on the mixture gas fuel equivalent ratio.

In flame combustion with CH4, the limit of flammability is determined around the fuel equivalence ratio 0.5 . But in this case, the reaction is conducted in the condition of the fuel equivalence ratio around 0.2 . With the sufficiently high preheating temperature and the fuel equivalence ratio 0.19 ~ 0.27 , the conversion rate achieved over 98% in this equipment. But, the unsteady operation was observed when the fuel equivalence ratio had the value outside 0.19 ~ 0.27. The conversion rate was investigated in the range of the fuel equivalence ratio from 0.16 to 0.3 in the condition of mixture gas flow rate 408.2 m3/h . Fig. 5 depicts the conversion rate in according to both the fuel equivalence ratio and the preheating temperature.

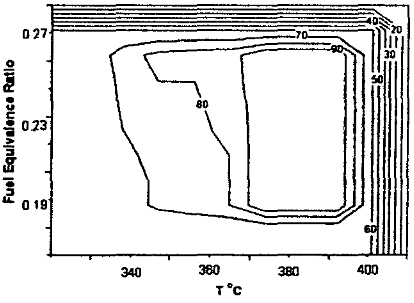

Fig. 5 The combustion characteristics on the fuel equivalence ratio of the mixture gas

In this catalytic combustor, the most steady operation was conducted around the fuel equivalence ratio 0.25 . This means that steady reaction has some range of mixture fuel equivalence ratio owing to the heat balance in the system. Both the overheating and the under-heating are a problem in the design of the catalytic application.

3.4 The combustion characteristics according to the temperature distribution of the catalytic honeycomb surface

The temperature distribution on the catalytic honeycomb surface got changed according to the ignition process of the catalytic combustor. Fig 6. shows the change of temperature distribution of catalytic front layer and rear one and the conversion rate according to the ignition process of catalytic combustor. In the case of preheating process using flame , the temperature of the catalytic honeycomb front layer which was radiated by the preheating flame was measured higher than that of rear one. Whereas, in the case of steady state which only catalytic combustion exists in , the temperature of the catalytic honeycomb rear layer was measured relatively high.

In the case of preheating, overall temperature of catalytic honeycomb layer is relatively high and then the conversion rate is almost 100%. After preheating , when the catalytic combustion began , the temperature distribution of both front and rear layer of the catalytic honeycomb got flatten. And the conversion rate was on the rise. When the catalytic combustion reached the steady state, the conversion rate come to 99%. At that time, the peak value of temperature of catalytic honeycomb layer is 50°C higher than in the case of preheating. Conclusively, as the more flatten temperature distribution of catalytic honeycomb layer got , the higher conversion rate got. And as the peak value of temperature of the honeycomb layer got higher, the conversion rate got more higher.

4. Conclusions

The catalytic heat exchanger was devised employing a regenerative preheating system for air. In this system, the mixture velocity did not affect significantly on the performance of catalytic combustor while the preheating temperature of air affected the conversion rate. The complete and stable conversion over the catalyzed honeycomb was achieved with preheated air of 370-390C, mixture velocity of 0.53 ~ 0.75 m/s and the lower equivalence ratio of 0.19 ~ 0.27. The catalytic heat exchanger device showed about 75 % heat exchange efficiency when air was used as a working fluid. Conclusively, both the heat balance in the system and the mixture condition determine to achieve steady state reaction of this system.

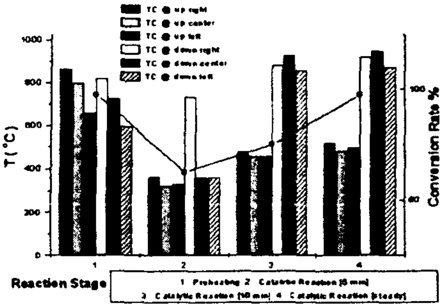

Fig. 6 The temperature distribution of the catalytic honeycomb layer and conversion rate according to the operational sequential process.

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