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Catalysis Today 26 (1995) 309–317

CATALYSIS TODAY

Multistage catalytic combustion systems and high temperature combustion systems using SiC

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Abstract

Multistage catalytic combustion systems for commercial applications using noble metal catalyst were investigated at conditions under 1000°C. Also, the high temperature combustion systems using SiC material were investigated at conditions up to 1400°C.

Keywords: Combustion; NO_x; Silicon carbide; Nitrogen oxides

1. Introduction

The catalytic combustion system is an advanced combustion technology having its characteristic feature in lower NO_x generation and is the most attractive combustion technology from the view point of earth environment problems. It is expected to apply to gas turbines and various industrial uses for NO_x emission control.

It is clearly desirable for thermal efficiency to completely consume all of the oxygen contained in the combustion air. But it is almost impossible in the conventional single-stage combustion systems because the combustion temperature will be extremely higher. So the combined systems of multistage catalytic combustion units will be effective to use oxygen completely. Concerning the application of multistage catalytic combustion, the combined power generation systems with a heating apparatus of reactor and a gas turbine have advantages.

This paper describes the commercial equipment with multistage catalytic combustion systems using precious metal catalysts under 1000°C and the high temperature combustion systems using SiC monolith at 1400°C.

2. Multistage catalytic combustion systems

Since catalytic combustion systems generally will be operated under fuel-lean conditions, the combustion gas from the first-stage of catalyst beds still contains enough oxygen to burn a fuel catalytically in the next stages. In addition, if the combustion gases are used as a heat resource for the start-up of each stage in heating equipment, the preheaters will be not required in each stage of catalyst beds except the first stage. On the other hand, the combustion temperatures in each stage are depending on the material properties and heating processes of equipment but it is easy to obtain the required combustion temperature by controlling the fuel/oxygen ratio.

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SSDI 0920-5861(95)00153-0

Therefore, the effective use of remained heat energy and oxygen in the combustion gases at each stage are characteristic advantages of the multistage catalytic combustion system. It leads to improve the total thermal efficiency of equipment. Consequently, to apply the multistage catalytic combustion system is quite advantageous for commercial combustion equipment as well as lower NO_x emission.

2.1. Heating systems with far-infrared radiation

A heating system for large spaces such as factory and housing area, which technology is extremely difficult so far, is effectively achieved by using far-infrared radiation. The first commercial products for baking and drying of coated paints of automobiles were established by Ford Motors, USA in 1938. Recently, far-infrared radiation has been frequently used for baking coated paints, sauna bath, food processing and heating systems in Japan too.

In electromagnetic waves, a variety of wave length is available as shown in Fig. 1. Far-infrared radiation is a part of the infrared radiation region having wave lengths between 3.5 μm and 1 mm. It closes to microwaves. For heating systems, far-infrared radiation having a wave length of about 5 μm is particularly efficient. Heat radiation from solid surfaces is generated by heating and the wave length changes are depending on the temperature of the solid surface. The relation between wave length and temperature of the solid surface is shown according to Wien's displacement law as follows [1]:

$$\lambda_{max} = 2897/T$$

where λ_{max} is the main wavelength (μm) and T the surface temperature (K). To generate far-infrared radiation, the temperature of the solid surface should be kept between 554°C and 17°C. The amount of radiation energy increases with increasing radiation rate. Accordingly, it is recommended to select ceramics having a higher radiation rate as radiation material.

In far-infrared radiators using catalytic combustion, the surface temperature of the radiation tube which generates far-infrared radiation of ca. 5 μm is needed to keep at ca. 300°C to Wien's displacement law. To keep the tube surface temperature at ca. 300°C, combustion gas of ca. 650°C must be supplied into the tube. It is very difficult to obtain combustion gas of ca. 650°C using a conventional diffusive combustor. But it is achieved by using a commercialized catalytic combustion system, which is able to make the gas with the required characteristic features.

In a multistage catalytic combustion system for a far-infrared radiator, an important feature required for the heating system is to give a uniform temperature any where in its installed area. Although it is advisable to keep the surface temperature of the radiation tube as even as possible, a substantial difference of temperature between

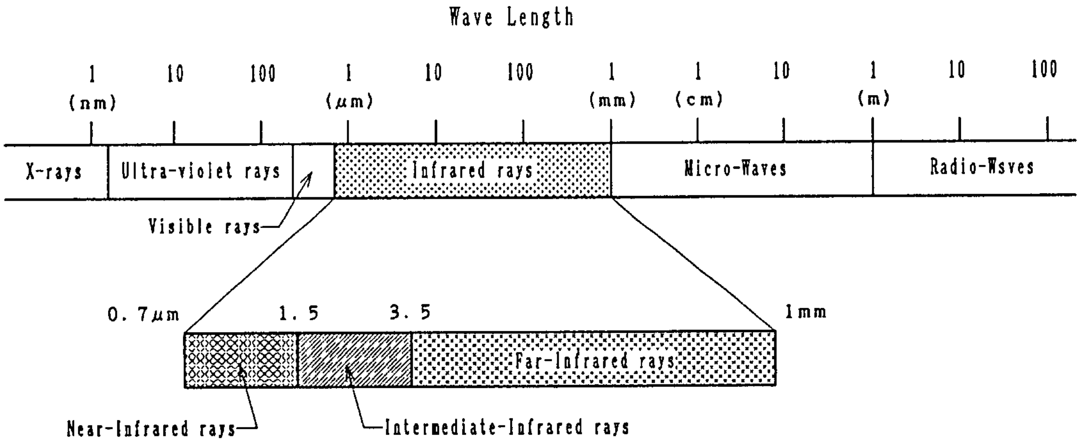


Fig. 1. Variety of electromagnetic waves.

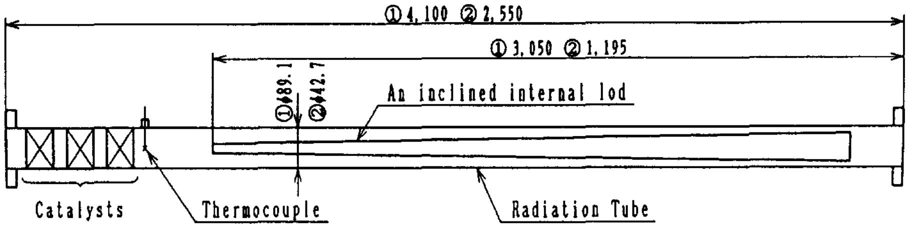


Fig. 2. Structure of radiation tube (1, commercial-type; 2, proto-type).

the inlet and outlet of tube cannot be avoided. Also, it is really impossible to extend the length of the radiation tube without any limitation. An example of a radiation tube with an inclined internal rod shown in Fig. 2 is a special device to obtain uniform surface temperatures of the radiation tube and the practical length of radiation tube is based on a theoretical calculation and experimental results.

In a large space, the combined heating system which consists of many heating apparatus with radiation tubes should be required. In such a system, the outlet gas of one radiation tube has still enough oxygen and temperature level to proceed catalytic combustion at the next stage. Catalytic combustion is progressing by feeding and mixing fuel into the outlet gas of the radiation tube. Theoretically, oxygen in the outlet gas will be completely consumed by the repetition of feeding and mixing fuel in the combined heating system with the multistage radiation tube system.

The superior characteristic feature of the multistage catalytic combustion system is the structure in which combustion apparatuses are connected in series with each other. It enables to use all heat energy effectively, because the exhausted gas is only discharged at the final stage of combustion [2,3].

2.2. Prototype equipment and experimental results

A prototype equipment based on Fig. 2 was manufactured for trial and the experiments were conducted before the optimum design of the commercial-type equipment. The schematic figure of the experimental apparatus is shown in Fig. 3. Combined catalysts of two monoliths of the M-type ceramic catalyst (210 cell/in^2, Pt 6 g/l) and one monolith of the F-type ceramic catalyst (400 cell/in^2, Pt 3 g/l + Pd 6 g/l) were used for one catalytic combustion unit in the experiments. Stainless steel tubes coated with a ceramic radiation material for far-infrared wave lengths were used as radiation tubes. The experimental condition is listed below and the results are shown in Fig. 4 and Fig. 5.

Number of catalytic combustion unit: 6 units
Height of radiation tube: 3.3 m
Air flow rate: 15 N m^3/h
Fuel flow rate: 1.3 l/h (kerosene)
Initial temperature of catalytic combustion: 200°C

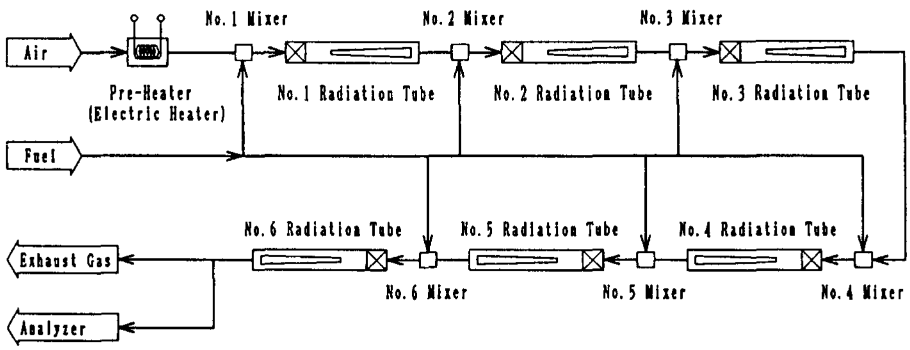
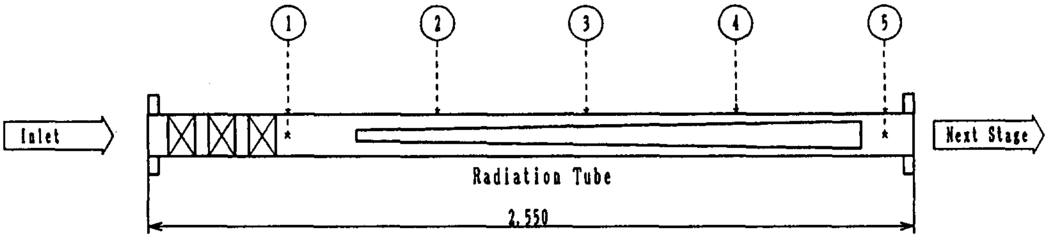


Fig. 3. Experimental flow.



Measuring Point		①	②	③	④	⑤
Temperrature	Inside of Tube (℃)	617				287
	Surface of Tube (℃)	360	264	247	247	231
Wave Length *	(μm)	4.6	5.4	5.6	5.6	5.8

* Calculated by Wien's Displacement Law

Fig. 4. Surface temperature of a radiation tube.

Remained oxygen concentration at the outlet of final stage: 1.6%

The experimental results on the prototype equipment suggested that the heating system with the multistage catalytic combustion unit could be efficient. However, to satisfy the requirement for commercial use, the following problems remain.

Since catalytic combustion uses an air-preheating system for each fuel, the electric preheater was used at the first-stage of the prototype equipment. In the commercial equipment, alternate preheating means should be developed because the continuous use of the electric heater will run the cost higher which is not recommendable. For that reason, a burner-type preheater was introduced for the development of the commercial equipment, instead of the electric heater. The preheater satisfies the requirements for low NO_x emission and compact structure [2,3].

2.3. Commercial equipment

Fig. 6 shows a low NO_x emission burner developed for a preheater of the commercial equipment which capacity is 20 000 kcal/h using propane and 700°C for the combustion gas temperature. The new burner without flame serves a more effective combustion system because of complete mixing by a venturi mixer. The operating results showed that the NO_x emission concentration is less than 30 ppm at the outlet of burner which is extremely lower than that of conventional burners.

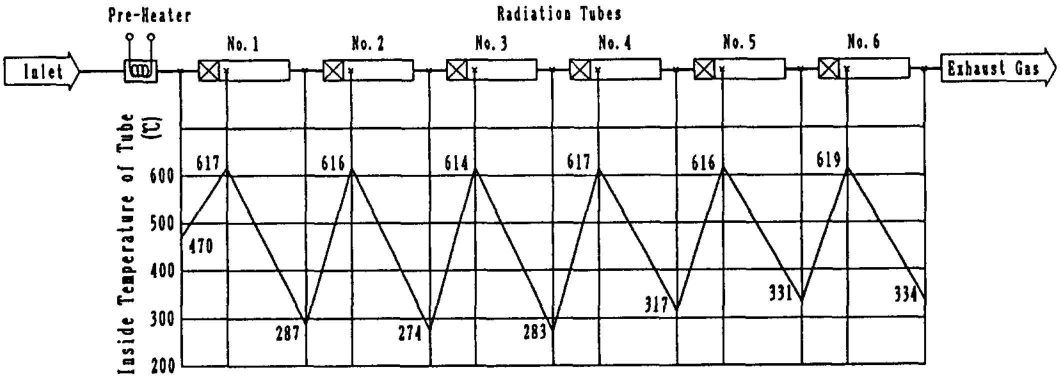


Fig. 5. Typical characteristics of combustion.

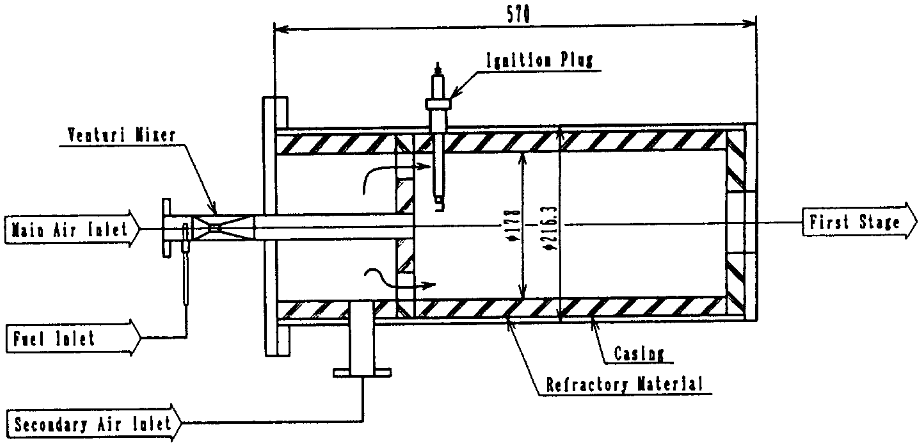


Fig. 6. A typical burner for preheater.

But still it is not enough for commercial heating systems in future.

In the commercial equipment, the low NO_x emission preheater is introduced and the flow system is almost similar to the system shown in Fig. 3. Fig. 7 shows a schematic flow system for commercial equipment. As a commercial plant, the multistage system is available for any installation place and it is advantageous to introduce the six-stage system because of its thermal efficiency.

An example of commercial operating conditions is listed below and the operating result is shown in Fig. 8 [2,3].

Number of catalytic combustion unit: 2 units
Height of radiation tube: 5 m
Fuel flow rate: 1.25 l/h (propane)
Air flow rate: 85 N m^3/h
Initial temperature of catalytic combustion: 300°C
Remained oxygen concentration at the outlet of the final stage: 13%

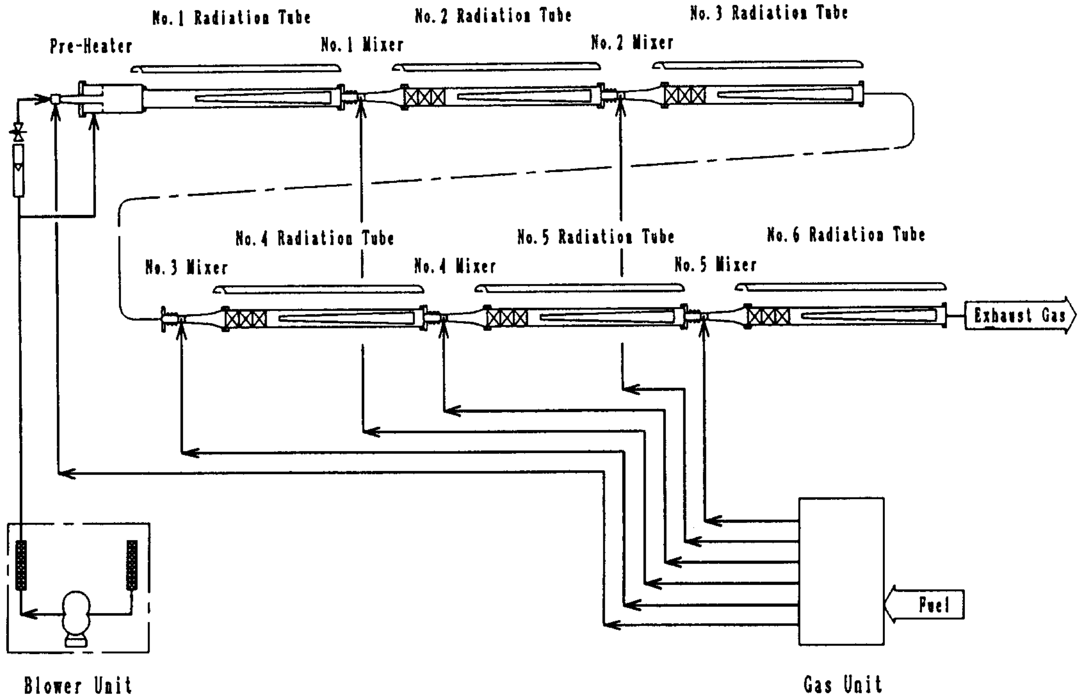
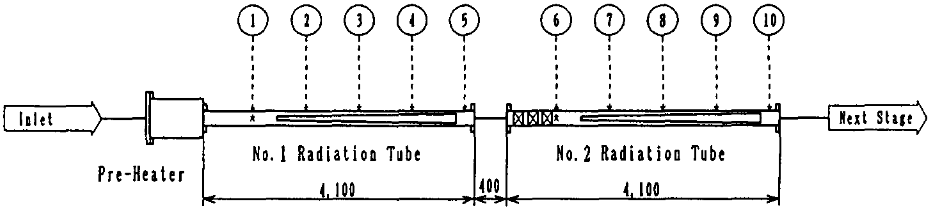


Fig. 7. Commercial type system flow.



Measuring Point		1	2	3	4	5	6	7	8	9	10
Temperrature	Inside of Tube (℃)	679					715				
	Surface of Tube (℃)	437	391	352	331	322	367	362	338	313	307
Wave Length * (μm)		4.1	4.4	4.6	4.8	4.9	4.5	4.6	4.7	4.9	5.0

* Calculated by Wien's Displacement Law

Fig. 8. Surface temperature of radiation tube.

2.4. *Summary of multistage catalytic combustion systems*

In addition to the heating systems, various types of clean and economical combustion systems can be developed by using the merits of catalytic combustion. Since preheated air is needed for the promotion of catalytic combustion, the development of a more improved preheater which is emission NO_x free is expected in near future.

On the other hand, another preheater of the catalytic combustion system for liquid fuels such as kerosene has also been developed and development of a new catalyst which initiates the catalytic combustion reaction at a minimal temperature should also be eagerly needed.

3. High temperature combustion systems using SiC

Recently, the development of low NO_x emission combustion equipment which will be operated at a higher temperature class of 1400°C was also needed. For the needs, high temperature combustion equipment which is a self-heating type with the tube and honeycomb monolith of SiC was developed. Fig. 9 shows a rough sketch of a single-tube high-temperature combustion equipment and Fig. 10 shows an example of typical experimental results of the NO_x emission of the single-tube system. Fig. 11 shows also a multi-tube high-temperature combustion equipment. This

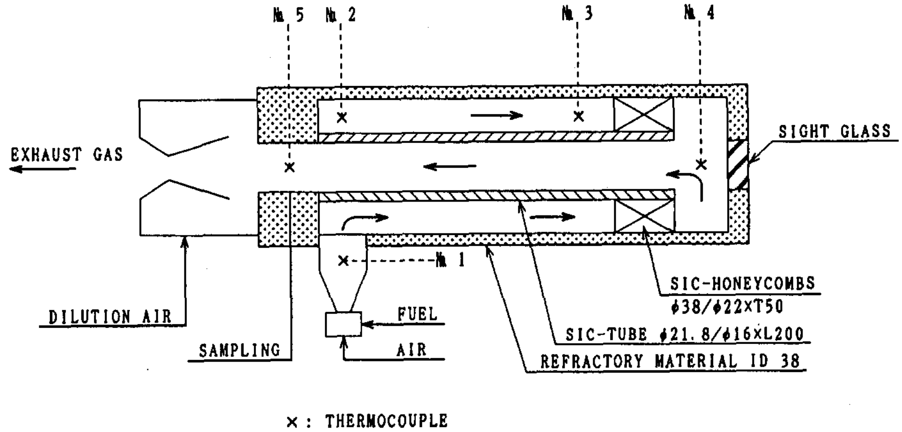


Fig. 9. Single-tube high-temperature combustion equipment.

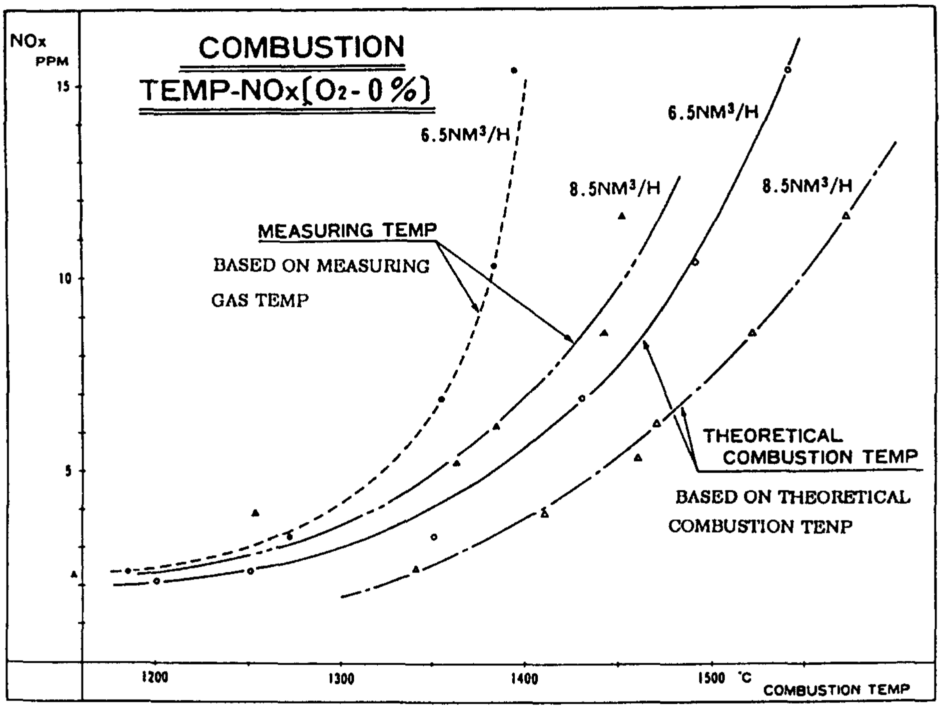


Fig. 10. Typical experimental data on NO_x emission using a single tube.

equipment will be operated by the following method [4,5].

At the start-up of the combustion equipment, the outside surface of the SiC tube was settled and the SiC honeycomb monolith was heated over 800°C which corresponded to an initial temperature of the reaction of 1200°C–1300°C of the combustion gas generated by the pilot burner. The combustion reaction was maintained by switching to the main fuel–air mixture from the primary fuel. After starting the main combustion, the pilot burner was shut off. The combustion gas was introduced into the inside SiC tube and heated the outside surface of the tube by heat conduction through the tube wall without the preheating of air for the gas fuel combustion. For liquid fuels, ca.

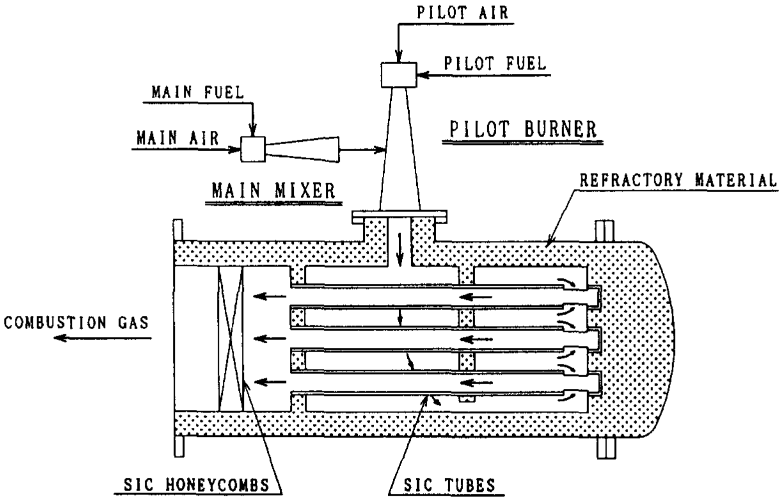


Fig. 11. Multi-tube high temperature combustion equipment.

200°C of preheating is necessary to mix the fuel and air well.

In these combustion systems, combustion temperatures are uniformed at any point. The temperature corresponded to the theoretical combustion temperature and the formation of NO_x was ultimately low.

It is considered that the reaction mechanism is as follows. Combustion of the fuel/air mixture is initiated by contacting with the surface of the SiC tube heated at the theoretical combustion temperature to generate a high temperature of the combustion gas. Successive chain combustion reactions occur by contacting with fresh feed fuel and combustion gas. In the combustion process, the chain reaction is promoted by the activation of fuel molecules which are activated at ca. 6 μm far-infrared radiation from the SiC tube surface and refractory materials.

The reasons why SiC was selected as a material for honeycomb monolith structures are listed below.

1. The durable temperature is extremely high (ca. 1400°C in air).
2. It has a superior thermal-shock resistance.
3. The thermal conductivity is fairly high.
4. The radiation efficiency is high.

The 110 thermal cycle tests were conducted for the thermal-shock resistance and durability of SiC by supplying a fuel/air mixture which a ratio corresponding to 1500°C in theoretical combustion temperature. The temperature pattern is shown in Fig. 12. As damages of the SiC tube and honeycomb monolith could not be observed as a result, the possibility of continuous use of SiC tube and honeycomb monolith up to 1400°C are confirmed.

In addition, multi-tube high temperature combustion equipment using the combination of SiC tubes and honeycomb monoliths are constructed recently and the system is operating now.

Its specification is listed below.

Fuel: kerosene
Operating pressure: 588–294 kPa
Combustion temperature: 1350°C

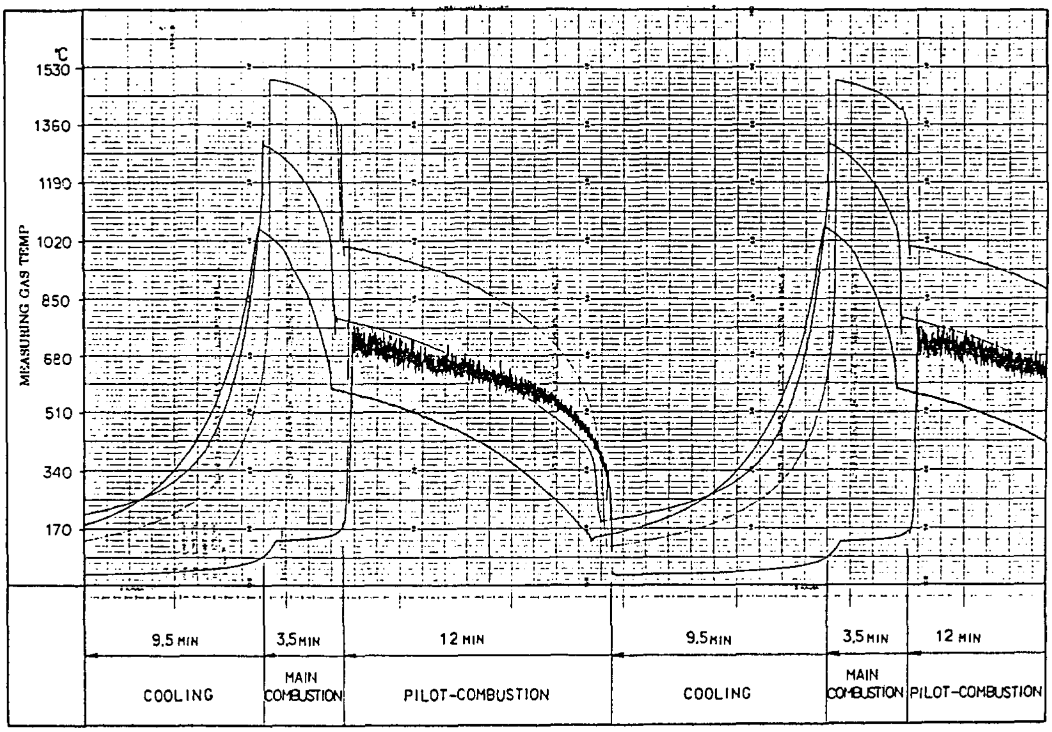


Fig. 12. Temperature pattern for the thermal cycle test of thermal shock resistance.

Heat generation: 4 Mkcal/h

4. Conclusion

The multistage catalytic combustion systems for the commercial applications using noble metal catalyst were investigated at conditions of under 1000°C. Also, the high temperature combustion systems using SiC material were investigated at conditions of up to 1400°C.

However, the following hard problems must be solved before the commercial use of multistage catalytic combustion systems for liquid fuels. First is the inevitable supply of preheated air for initiating the first-stage of catalytic combustion. Second is the heating method of SiC for the starting reaction after the second-stage combustion. Our efforts should be concentrated onto such a development to achieve a breakthrough for completion of advanced multistage catalytic combustion systems.

Acknowledgements

The authors thank Dr. H. Fukuzawa for valuable advice on combustion systems development over the past few years.

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