ELSEVIER

Catalysis Today 47 (1999) 399–405

CATALYSIS TODAY

Development of a catalytically assisted combustor for a gas turbine[1]

Yasushi Ozawa[a,*], Tomoharu Fujii[a], Mikio Sato[a], Takaaki Kanazawa[b], Hitoshi Inoue[b]

[a] *Central Research Institute of Electric Power Industry, 2-6-1 Nagasaka, Yokosuka, Kanagawa 240-01, Japan*
[b] *Kansai Electric Power Company, Inc., 3-11-20 Nakoji, Amagasaki, Hyogo 661, Japan*

Abstract

A catalytically assisted low NO_x combustor has been developed which has the advantage of catalyst durability. This combustor is composed of a burner section and a premixed combustion section behind the burner section. The burner system consists of six catalytic combustor segments and six premixing nozzles, which are arranged alternately and in parallel. Fuel flow rate for the catalysts and the premixing nozzles are controlled independently. The catalytic combustion temperature is maintained under 1000°C, additional premixed gas is injected from the premixing nozzles into the catalytic combustion gas, and lean premixed combustion at 1300°C is carried out in the premixed combustion section. This system was designed to avoid catalytic deactivation at high temperature and thermal or mechanical shock fracture of the honeycomb monolith. In order to maintain the catalyst temperature under 1000°C, the combustion characteristics of catalysts at high pressure were investigated using a bench scale reactor and an improved catalyst was selected for the combustor test. A combustor for a 20 MW class multi-can type gas turbine was designed and tested under high pressure conditions using LNG fuel. Measurements of NO_x, CO and unburned hydrocarbon were made and other measurements were made to evaluate combustor performance under various combustion temperatures and pressures. As a result of the tests, it was proved that NO_x emission was lower than 10 ppm converted at 16% O_2, combustion efficiency was almost 100% at 1300°C of combustor outlet temperature and 13.5 ata of combustor inlet pressure. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Catalytic combustion; Gas turbine; Natural gas; Nitrogen oxides

1. Introduction

Gas turbine combined cycle power plants has been used widely in recent years because of their advantage in thermal efficiency. However, the regulatory requirements for low NO_x emissions from gas turbine plants have also been made stricter. Catalytic combustion is an extremely effective method to reduce NO_x emissions [1], but because of catalyst deactivation at high temperatures, catalytic combustors for gas turbines have not been commercialized.

With the aim of overcoming this problem and achieving the combustion performance indicated in Table 1, joint R&D on low NO_x combustor applied catalytic combustion was started by Central Research Institute of Electric Power Industry (CRIEPI) and Kansai Electric Power Co. Inc. (KEPCO) in 1988 [2]. In this work, a catalytically assisted, premixed combustor was designed and an atmospheric test was

---

*Corresponding author. Tel.: +81-468-562121; fax: +81-468-563346; e-mail: ozaway@criepi.denken.or.jp

[1] Presented at the 3rd International Workshop on Catalytic Combustion, Amsterdam, The Netherlands.

PII: S0920-5861(98)00322-8

Table 1
Targets for combustor performace

Combustor exit gas temperature (T_g)	1300°C
NO_x emission	<10 ppm
Combustion efficiency (η)	>99.9%
Total pressure loss(ΔP)	<5%
Pattern factor (PF)	<15%

completed in 1993 [3]. At the same time, the catalyst was improved. Then the combustor was tested at high pressure conditions.

2. Design of combustor

The difficulties of applying combustor to a gas turbine are as follows:

- Thermal degradation of the catalyst
- Thermal shock fracture of the ceramic monolith of the catalyst
- Necessity of a uniform fuel/air mixture

In order to overcome the difficulties, the catalytically assisted, premixed combustor shown in Fig. 1 was designed.

The scale is equivalent to one combustor of a 20 MW class multi-can type gas turbine. The combustor is composed of a burner section and a premixed combustion section. The burner section shown in Fig. 2 consists of an annular preburner, six catalytic combustor segments and six premixing nozzles. The premixed combustion section consists of a metal wall liner and a transition piece. The catalyst segments and premixing nozzles are arranged alternately to form a circle.

Air is heated to 370°C by the preburner and is distributed to the catalyst segments and premixing nozzles. Catalytic combustion is conducted below 1000°C and premixed gas from the premixing nozzles is injected into the catalytic combustion gas at right angles, then re-circulating flow occurs after the end face of the premixing nozzles. As a result, stabilized by both the catalytic combustion gas and the re-circulating flow, lean premixed combustion at 1300°C is carried out.

In this design, because small catalysts are able to be applied to the combustor, the structural reliability of the ceramic monolith of catalyst will be maintained. Moreover, because of keeping the catalyst temperature low, deterioration of the catalysts caused by heat will

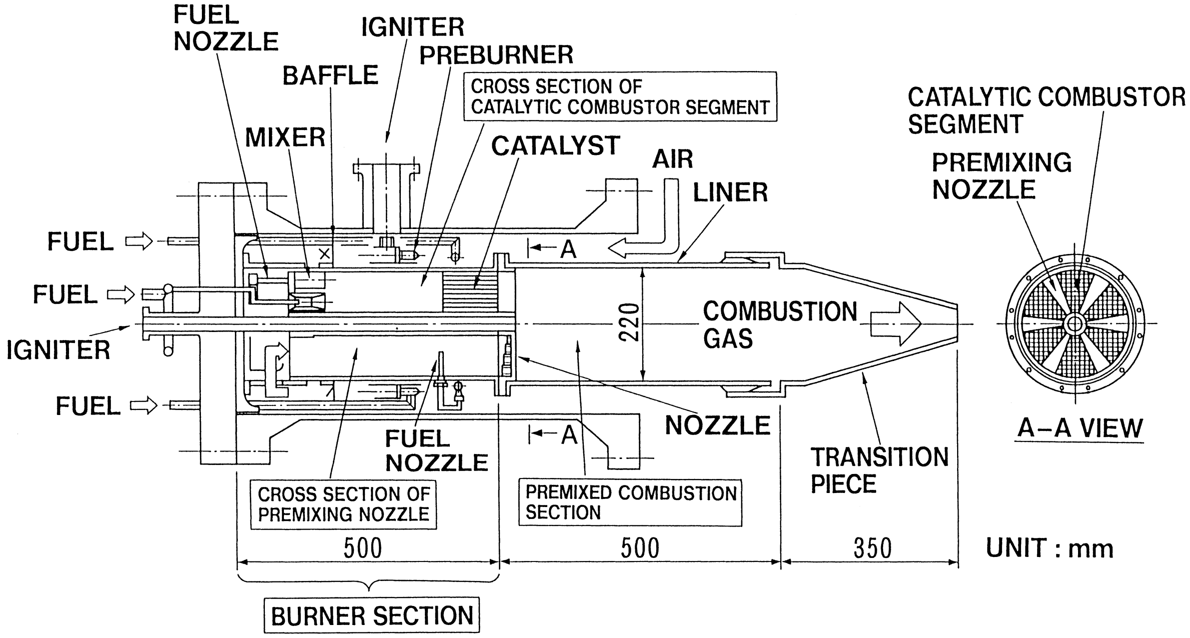

Fig. 1. Schematic figure of the combustor.

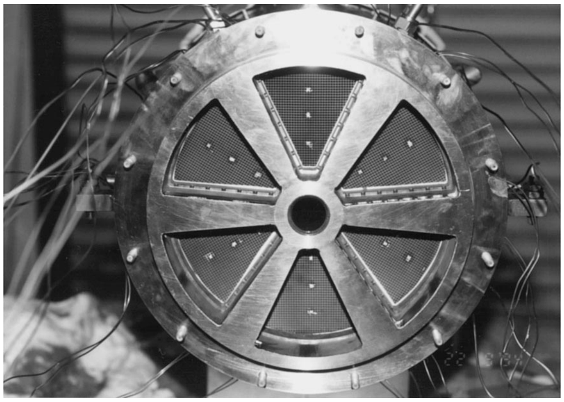

Fig. 2. Burner section.

be prevented and a certain degree of lack of uniformity in the fuel/air mixture is permitted.

3. Catalyst improvement

In the case of this combustor, NO_x emission is mainly caused by the premixed gas from the premixing nozzles, which has a higher fuel concentration than that for the catalytic combustor segments. For this reason, it is necessary to make the fuel concentration for the catalytic combustor segments as high as possible to reduce NO_x emission while keeping the catalyst temperature under 1000°C. Another way to reduce NO_x emission is to reduce the ignition temperature of the catalyst to diminish the load of the preburner. From these standpoints, catalyst improvement was studied.

3.1. Experiments

Table 2 shows the properties of a base catalyst of Pd/Pt and an improved catalyst of Pd/Pt/Rh. In the case of methane combustion by the Pd/Pt catalyst, self-oscillation phenomena were observed, which became an obstacle to the fuel increase of the catalyst. It was suggested by Inui et al. [4] that Rh holds the possibility of reducing self-oscillation, so that a small amount of Rh was added. In order to compare the aging effect of the catalysts, the samples were aged in air within an electric furnace for 1000 h. The aging temperature was kept at 750°C, which was the mean temperature of the catalysts in catalytic combustion.

Table 2
Catalyst properties

	Pd/Pt	Pd/Pt/Rh
Substrate	Cordierite honeycomb monolith ϕ25 mm×L25 mm×200 cells/in^2	
Washcoat	Stabilized Al_2O_3 80 g/l+ZrO_2 20 g/l	
Catalyst	Pd 20 g/l+Pt 5 g/l	Pd 20 g/l+Pt 5 g/l+ Rh 2 g/l

Table 3
Test conditions

Pressure (P)	9 ata
Space velocity (SV)	7.8×10^6 h^{-1}
Excess air ratio (λ)	2.9
Fuel	Natural gas (CH_4=99.2%, CO_2=0.7%, N_2=0.1%)

At first, the ignition temperature was examined under the conditions in Table 3. The rising rate of the catalyst inlet temperature (T_{ci}) was kept at 2°C/min. The ignition temperature was defined as the T_{ci} at the point of rapid increase of the outlet gas temperature of the catalyst.

Next, keeping the catalyst inlet temperature at 400°C and the other conditions constant, the reaction temperature was observed for 7 h.

3.2. Results

Fig. 3 shows the temperature. For the fresh catalysts, the ignition activity of Pd/Pt/Rh was similar to that of Pd/Pt, but after aging for 1000 h, it was better

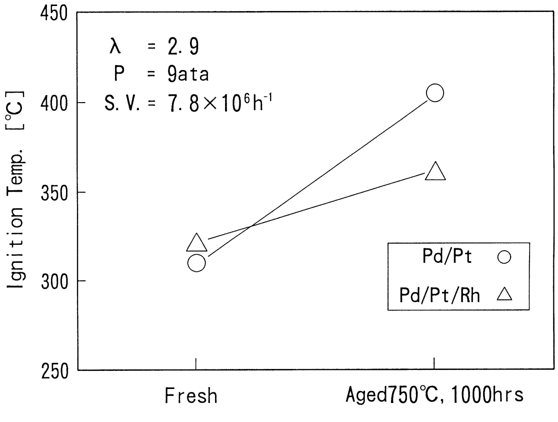

Fig. 3. Ignition temperature

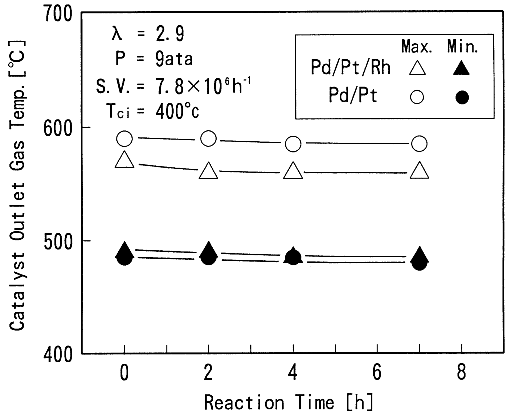

Fig. 4. Trend of reaction temperature of fresh catalyst.

than that of Pd/Pt and the ignition temperature was under 370°C, which was the reference inlet temperature for the combustor under full load conditions.

Fig. 4 shows the trend of the outlet gas temperature for fresh samples under constant conditions. The temperature amplitude of Pd/Pt/Rh was smaller than that of Pd/Pt. Fig. 5 shows the trend of the outlet gas temperature of aged catalysts for 1000 h. Compared with the results of fresh samples, there was little activity deterioration of both types of catalysts by aging and little influence of aging on the temperature amplitude.

By the addition of Rh, the ignition temperature was lowered. And as self-oscillation was reduced, it was considered to be possible to increase the fuel concentration of the catalyst under the upper limitation of the catalyst temperature.

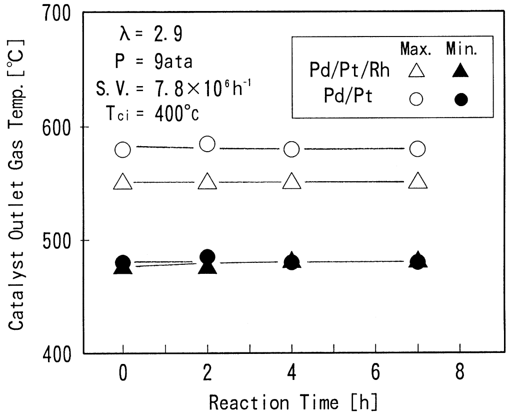

Fig. 5. Trend of reaction temperature of aged catalyst at 750°C, 1000 h.

4. Combustor test

4.1. Test facility and conditions

Fig. 6 shows a schematic diagram of the test facility for a gas turbine combustor. This facility can test one combustor for 20 MW class multi-can type gas turbine in a pressure range from 3 to 13.5 ata and in an air flow rate range from 3000 to 13 500 m^3 N/h.

Air from a compressor is heated by an air heater and fed through a flow control valve to a vessel in which the combustor was included. Air to the vessel is partially used to cool the liner and transition piece of the combustor and remaining air is introduced into the burner section. Air to the burner section is distributed to catalytic combustion segments and premixing nozzles. Fuel is injected into mixers in the catalytic combustor segments, lean premixed gas is provided to the catalyst segments. Subsequently, fuel is injected into the premixing nozzles and premixed gas is injected from the nozzles into the catalytic combustion gas. For smooth ignition of the premixed gas, an igniter is inserted in the premixed combustion section. In the measurement duct which is behind the combustor outlet, the combustion gas temperature is measured by thermo-couples and emission is sampled by a sampling probe. Emission is analyzed by the methods shown in Table 4, and combustion efficiencies are calculated.

Table 5 shows the fuel properties, which are sampled from the fuel line and analyzed by TCD gas chromatography. Fuel is fed to three lines of preburner, catalyst and premixing nozzles, so that the loads of the respective combustion sections can be controlled by the respective fuel flow rates.

Table 4
Analyzing method

Composition	Analyzing method
NO_x	Chemi luminescence
CO	Non-dispersive infrared
CO_2	Non-dispersive infrared
O_2	Paramagnetic
Unburned hydrocarbon (UHC)	Flame ionization

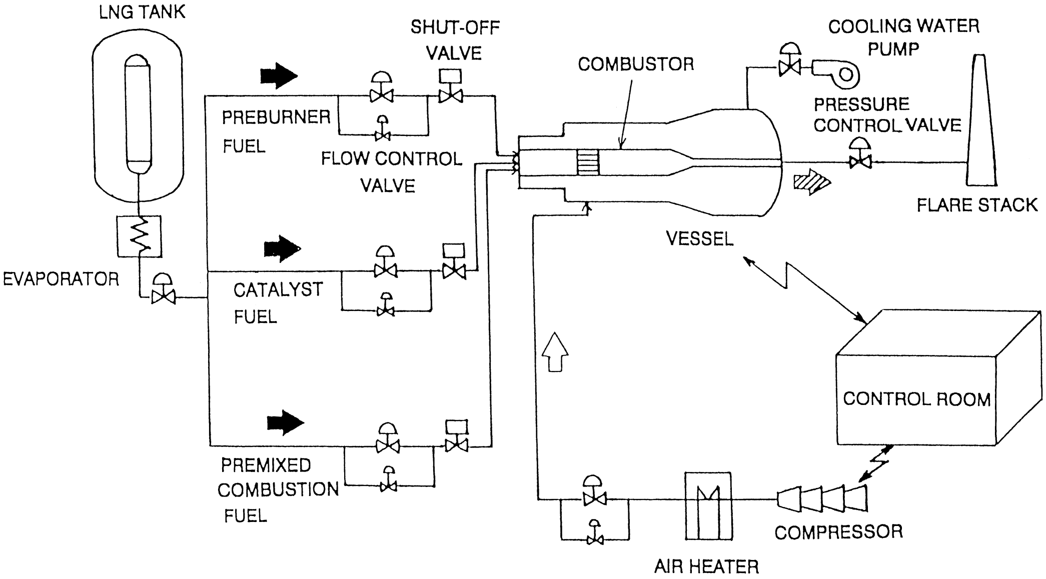

Fig. 6. Schematic diagram of test facility.

Table 5
Fuel compositions

CH_4	87.65%
C_2H_6	4.64%
C_3H_8	4.23%
i-C_4H_{10}	1.55%
n-C_4H_{10}	1.75%
i-C_5H_{12}	0.15%
n-C_5H_{12}	0.00%
N_2	0.03%

An external view of the catalyst segments is shown in Fig. 7. The catalyst is Pd/Pt/Rh. The size is 25 mm in length and 200 cells/in^2 and has six fan-shaped segments formed by dividing a ring of 40 mm i.d. and 200 mm o.d. and arranged in two stages. The catalyst temperatures are measured by thermo-couples which are inserted and cemented in the catalyst cells.

Table 6 shows the conditions of a full load. In this test, the preburner was not ignited but a small amount of fuel was injected from the preburner as though it was a fuel nozzle for the purpose of promoting the mixing of fuel with air. In order to keep the catalyst inlet temperature at 370°C, the combustor inlet temperature needed to be maintained at 400°C because of the radiation of heat from the vessel. Catalytic combustion and premixed combustion was initiated under the conditions of 3 ata and 3000 m^3 N/h of air. When ignition had been completed, the pressure, air flow rate and fuel rate were increased to the full load gradually.

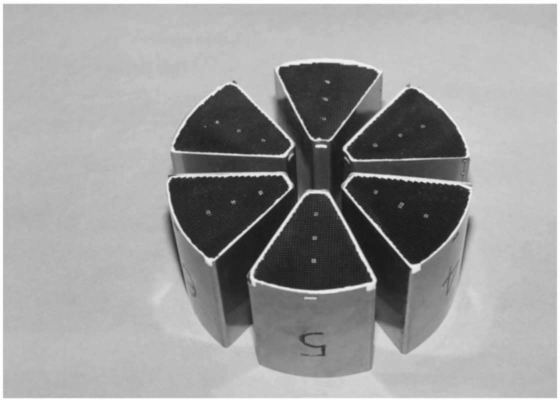

Fig. 7. Catalyst segments.

4.2. Results

Fig. 8 shows the relation between pressure and emissions. The pressure was increased from a low pressure while keeping an excess air ratio (λ) of 2.6. Assuming the application of an air bypass valve [5], the air flow rate was increased in proportion to the pressure. The catalyst inlet gas temperature and the

Table 6
Standard test conditions

Pressure (P)	13.5 ata
Total air flow rate (V_a)	13 500 m^3N/h
Catalyst Inlet gas temperature (T_{ci})	370°C
Catalyst temperature (T_c)	<1000°C
Combustor outlet gas temperature (T_g)	1300°C

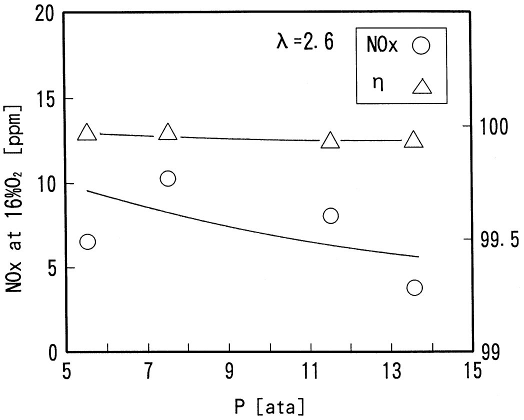

Fig. 8. Effect of pressure.

catalyst temperature were maintained at 370°C and about 750°C–800°C respectively.

Combustion efficiency (η) was almost 100% and stable combustion was kept between 3.5 and 13.5 ata. On the other hand, through the test, it was necessary to keep the catalyst temperature over 650°C to maintain stable premixed combustion. In a combustion system in which no fuel was added to the catalytic combustion gas, it was necessary to raise the catalyst temperature to about 900°C for the post catalyst combustion [6]. In contrast, because a richer premixed gas was injected in the catalytic combustion gas and a re-circulating flow occurred after the burner section, it was supposed that the stable premixed combustion was kept at such a low catalyst temperature in this combustor.

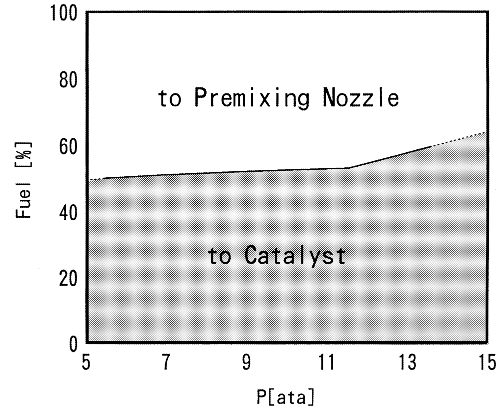

Fig. 9. Fuel distribution.

The maximum NO_x emission was 10 ppm converted at 16% O_2. NO_x emissions tend to decrease with increasing pressure. Fig. 9 shows the trend of fuel distribution to the catalysts and the premixing nozzles. In this figure, fuel to the preburner was shared to the catalysts and the premixing nozzles according to the air distribution, which was calculated by the fuel concentration in the premixed gas of each section. Because the fuel conversion in the catalyst bed decreases with increasing pressure, fuel distribution of the catalyst increased with increasing pressure in order to keep the catalyst temperature constant, and that to the premixing nozzles decreased. Thermal NO_x is formed in high temperature zones, which was caused by a fuel concentration of premixed gas from the premixing nozzles which is higher than that of the catalysts. For these reasons, NO_x emission is surmised to have been decreased with the rise of pressure.

Fig. 10 shows the effect of the combustor outlet gas temperature (T_g) on the combustion characteristics at 13.5 ata. In this test, the fuel for the preburner and the catalyst was kept constant and the fuel for the premixing nozzles was increased gradually to raise the combustor outlet gas temperature from 1150°C to 1300°C.

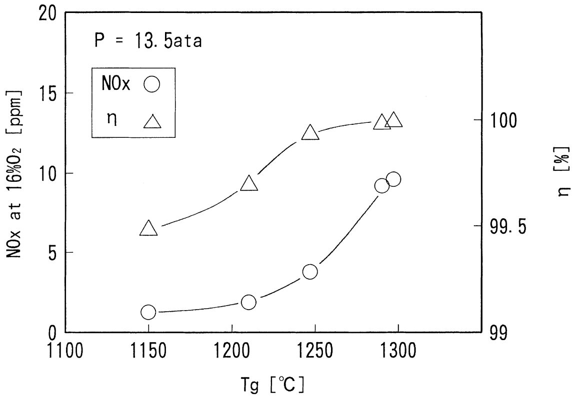

Fig. 10. Effect of combustion temperature.

The NO_x emission was increased with the rise of combustor outlet gas temperature, but was kept below 10 ppm (at 16% O_2) at a combustor outlet gas temperature of 1300°C. Combustion efficiency (η) was increased with increasing combustor outlet gas temperature, and was over 99.9% at 1250°C.

Pressure loss was almost constant at any pressure. The maximum overall combustor pressure loss was about 2.5%, and satisfactorily lower than the targeted level of 5%. Pattern factor, which is the index of the temperature uniformity of combustor outlet gas, was not measured.

In the atmospheric pressure combustion test of this type of combustor [3], NO_x emission was under 2 ppm (at 16% O_2), which was lower than the result of this test. These differences are mainly due to the structure of the combustion liners. A ceramic fiber type liner [7] was applied in the atmospheric test. When using the ceramic fiber type liner, the air distribution for the liner cooling can be decreased and the air to the premixing nozzles increased. As a result, the fuel concentration in the premixing nozzles decreased and it will be possible to lower the NO_x emission.

5. Conclusion

A catalytically assisted combustor was designed. The catalyst was improved for this combustor and the combustor for a 20 MW class gas turbine was tested at high pressures. As a result, NO_x emission was below 10 ppm at a combustor outlet gas temperature of 1300°C. It was supposed to be possible to lower NO_x emission by decreasing the liner cooling air. Based on these results, further work will be conducted on this combustor applied ceramic liner under high pressure conditions.

Acknowledgements

Our deepest gratitude goes to T. Iuni of Kyoto University who directed in the catalyst improvement, S. Kikumoto, T. Yamanaka and J. Hirano of KEPCO and Y. Tochihara of CRIEPI who joined this work, T. Hisamatsu and T. Abe of CRIEPI who helped us in promoting this work, H. Fukuzawa who helped us in catalyst development, S. Mandai of Mitsubishi Heavy Industry for the metal liner fabrication and the personnel of Tanaka Kikinzoku Kogyo for the catalyst fabrication.

References

[1] W.C. Pfefferle, R.V. Carrubba, R.M. Heck, G.W. Roberts, ASME Paper No. 75-WA/FU-1.
[2] Y. Ozawa, M. Saiga, M. Sato, S. Watanabe, M. Okahata, 91-YOKOHAMA-IGTC-107.
[3] Y. Ozawa, T. Fujii, S. Kikumoto, M. Sato, H. Fukuzawa, M. Saiga, S. Watanabe, Catal. Today 6 (1995) 351.
[4] T. Inui, T. Twana, Studies Sur. Sci. Catal. 19 (1984) 205.
[5] P.W. Pillsbury, K.L. Rieke, J.L. Toof, E.J. Wisniewski, EPRI Rep. AP-2584.
[6] R.A. Dalla Betta, J.C. Schlatter, S.G. Nickolas, D.K. Yee, T. Shoji, ASME Paper No. 94-GT-260.
[7] T. Hisamatsu, T. Abe, CRIEPI Rep. EW88004, 1989.