Proceedings of the International Gas Turbine Congress 2003 Tokyo
November 2-7, 2003

IGTC2003Tokyo TS-144

Effect of Primary Equivalence Ratio on Reducing both Fuel-NOx and Thermal-NOx Emissions of the Gas Turbine Combustor for Oxygen-blown IGCC with Hot/Dry Syngas Cleanup

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ABSTRACT

In order to improve the thermal efficiency of the oxygen-blown IGCC (Integrated Gasification Combined Cycle) and to meet stricter environmental restrictions among cost-effective options, a hot/dry synthetic gas cleanup is one of the most hopeful choices. The flame temperature of medium-Btu gasified fuel used in this system is high so that NOx formation by nitrogen fixation results to increase significantly. Additionally, the gasified fuel contains fuel nitrogen, as ammonia, and it produces nitrogen oxides, the fuel NOx, in the case of employing the hot/dry synthetic gas cleanup. Low NOx combustion technology to reduce both fuel-NOx and thermal-NOx emissions has been required to protect the environment and ensure low cost operations for all kinds of oxygen-blown IGCC. In this paper, we have investigated effects of the two-stage combustion for reducing both fuel-NOx and thermal-NOx emissions, and clarified the conditions for practical application. And also we tested a combustor, examined the effects of two-staged combustion with direct injection of nitrogen produced from air separation unit, and showed the engineering guidelines for the low-NOx combustor design of oxygen-blown gasified, medium-Btu fuels.

NOMENCLATURE

C.R. : conversion rate from ammonia to NOx %
D : inner diameter of the combustor mm
HHV : higher heating value of the fuel MJ/m^3
LHV : lower heating value of the fuel MJ/m^3
Ic : combustion intensity in the combustor W/(m^3 ·Pa)
N_2/Fuel : nitrogen over fuel supply ratio kg/kg
N_2total : total supply rate of nitrogen kg/s
N_2(BY) : flow rate of bypassing nitrogen premixed with the combustion air kg/s
NOx（16%O_2）:NOx emissions corrected at 16% oxygen in the exhaust gas ppm
NOx_{th} : thermal NOx emissions ppm
P : pressure inside the combustor MPa
T_{air} : air inlet temperature K
T_{ex} : average temperature of combustor exit gas K
T_{fuel} : fuel inlet temperature K
TN_2 : nitrogen inlet temperature K
Ur : mean velocity of cross-sectional flow of air (at 0℃ basis) m/s
θ : angle of fuel injection nozzle degree
ϕ_{ex} : equivalence ratio at combustor exit
ϕ_p : equivalence ratio in the primary combustion zone
ΔP/q : total pressure loss coefficient (characteristics section is combustor-exit)

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Manuscript Received on May 7, 2003

INTRODUCTION

IGCC is considered one of the most important systems for future coal utilization technology in power generation systems, and is being promoted throughout the world. In Japan, the government and electric power companies undertook experimental research at a 200T/D pilot plant project (Ichikawa, 1996) from 1986 to 1996. The Central Research Institute of Electric Power Industry (CRIEPI) developed an air-blown pressurized two-stage entrained-flow coal gasifier (Kurimura, 1995), a hot/dry synthetic gas cleanup system (Nakayama, 1990), 150MW, 1573K-class (Nakata, 1993) and 1773K (1500℃)-class gas turbine combustor technologies for low-Btu fuel (Hasegawa, 1998a). Of late, the government and electric power companies are promoting a demonstration IGCC project.

Other developments concerning the IGCC system and gas turbine combustor using the oxygen-blown gasified coal fuel include: The Cool Water Coal Gasification Project (Savelli, 1985), the flagship demonstration plant of IGCC; the Shell process (SGCP) (Bush, 1991) in Buggenum as the first commercial plant, which started test operation in 1994 with commercial operation from 1998; the Wabash River coal gasification repowering plant (Roll, 1995) in the United States, in operation since 1995; the Texaco process at the Tampa power station (Jenkins, 1995), in commercial operation since 1996; a HYCOL gasification process for the purpose of hydrogen production, which was developed in Japan (Ueda, 1995); and IGFC (Integrated coal gasification fuel cell combined cycle) pilot plant which consists of gasifier, fuel cell generating unit and gas turbine, in test operation from 2002 by Electric Power Development Co., Ltd. in Japan. Furthermore, the diversification of fuels used for the electric power industry, such as biomass, poor quality coal and residual oil, are also the most significant issues for gas turbine development in IGCC: The development of biomass-fueled gasification received considerable attention in the United States in the early 1980s (Kelleher, 1985) and the prospects for commercialization technology appear considerably improved at present (Consonni, 1997); Our research institute has started researching into the gasification technology of orimulsion^{TM} (emulsion of Orinoco tar) fuel (Ashizawa, 1996). All of the systems, which used oxygen as an oxidizer, were assumed to adopt the wet type synthetic gas cleanup system. Moreover, in almost all systems, premixing the surplus nitrogen, produced from the oxygen production unit, with a gasified fuel, increases gas turbine output and suppresses NOx emissions. From the viewpoint of both high operating costs and initial costs of removing the NOx in exhaust gas derived from the gas turbine system, the electric power industry aims for low-NOx combustion technology that promises higher thermal efficiency and environmentally-sound options.

With respect to the research into low-NOx combustion technology using medium-Btu gaseous fuel, other studies include: White et al. (1983) studied on the rich-lean combustor for low- and medium-Btu gaseous fuels; Dobbeling et al. (1994) studied on low NOx combustion technology which quickly mixed fuel with air using the ABB double cone burner (called EV burner); Dobbeling et al. (1996) studied on the premixed combustion characteristics of medium-Btu gaseous fuel in a fundamental small burner for low NOx emissions, because the burning velocity of medium-Btu fuel was about 6 times greater than conventional natural gas, a premixed combustion for low NOx emissions was so far difficult to adopt; Cook et al. (1994) studied on the effective method of returning nitrogen to the cycle, where nitrogen is injected from the head end of the combustor for NOx control; Zanello and Tasselli (1996) studied on the effects of steam content in the medium-Btu gaseous fuel on combustion characteristics; Hasegawa et al. studied on low-NOx combustion technology using surplus nitrogen injected from the burner (1998b, 1999a) and with lean combustion of instantaneous mixing (2003). Furthermore, we have been developing the low-NOx combustion technology for reducing both fuel-NOx and thermal-NOx emissions, in the case of employing the hot/dry synthetic gas cleanup to oxygen-blown IGCC (2002).

This paper will show the effectiveness of the two-stage combustion with surplus nitrogen direct injection from the burner and also the influences of the fuel composition, velocity of combustion gas and pressure inside a combustor on the NOx emission characteristics.

DESCRIPTION OF THE OXYGEN-BLOWN IGCC SYSTEM

Characteristics of the Oxygen-Blown IGCC System

In the oxygen-blown IGCC system, large quantity of nitrogen is produced in the air separation unit. In almost all of the systems, gasified coal fuels, premixed with the rest of the nitrogen, a part of which is used to feed coal into the gasifier, etc., are injected into the combustor to increase electric power and to decrease thermal-NOx emissions from the gas turbine. In this paper, we will develop a combustor for oxygen-blown type gasification system with a hot/dry synthetic gas cleanup facility, in which coal is fed into the gasifier with some water by slurry state or with some nitrogen by pulverulent body state, and the rest of the nitrogen is directly injected into the gas turbine combustor. The development of the combustor is intended for 1773K (1500℃)-class gas turbines, in which gasified fuels contain ammonia not removed in the hot/dry gas cleanup. Furthermore, it is necessary to return a large quantity of nitrogen produced from the air-separation unit (as much as the fuel flow rate) to the cycle from the standpoint of recovering power for oxygen production. Basically, the flow rate of the surplus nitrogen produced in the air-separation unit is almost proportional to the fuel flow rate at any gas turbine load, and all surplus nitrogen should be effectively injected into a gas turbine combustor prior to a turbine.

Characteristics of Oxygen-Blown Gasified Fuel

The typical compositions of medium-Btu gasified fuels produced in oxygen-blown gasifiers are shown in Table 1 (Hasegawa, 2003). Each gasified fuel contains carbon monoxide (CO) and hydrogen (H_2) as the main combustible components, a small fraction of methane (CH_4) and so on. Fuel calorific values vary widely (5-13MJ/m^3), from about one-eighth to one-third of natural gas, with raw materials and gasifier types. For example, a gasified fuel derived from biomass contains 30-40% steam in the fuel.

In the case of gasified coal fuel, the theoretical adiabatic flame temperature rises as the fuel calorific value increases. Fuel calorific values of 4.2MJ/m^3 and 12.7MJ/m^3 produce maximum flame temperatures of 2,050K and 2,530K respectively. When the fuel calorific value is 8.4MJ/m^3 or higher, the maximum flame temperature of the medium-Btu fuel without nitrogen is about 400K higher than that of the nitrogen-blended fuel (Hasegawa, 2003). That is, the flame temperature of medium-Btu gasified fuel, produced in an oxygen-blown gasifier, is higher than that of high-calorie gases such as natural gas which consists mainly of methane, while the medium-Btu fuel has a calorific value as low as one fifth of methane. Thermal-NOx emissions are expected to increase more when burning medium-Btu fuel than burning high-calorie gas of natural gas. We intend to inject surplus nitrogen directly into higher temperature regions from the burner and to decrease thermal-NOx emissions produced from these regions effectively.

Furthermore, gasified fuels contain ammonia when the hot/dry synthetic gas cleanup is employed. Development of low NOx combustion technology is necessary in order to reduce fuel-NOx emissions originating from ammonia in the fuel at the same time as reducing thermal-NOx emissions.

DESIGN OF THE COMBUSTOR

Problems of the Medium-Btu Fueled Combustor

From the characteristic of medium-Btu, gasified fuel as mentioned above, it could be said that the design of a gas turbine combustor, utilizing nitrogen supply into a combustor, should consider the following issues for an oxygen-blown IGCC with the hot/dry synthetic gas cleanup:

(1) Low NOx-emission technology: Thermal-NOx production from nitrogen fixation using nitrogen injection into the combustor, and fuel-NOx emissions originating from ammonia using a two-stage combustion must be simultaneously restrained.

(2) Higher thermal efficiency: Nitrogen injection must be tailored so as to decrease the power to compress nitrogen, which is returned into the gas turbine in order to recover a part of the power used for the air-separation unit.

Test Device and Methods for Basic Studies

Figure 1 shows an outline of the combustor and the tested burner. The combustor consists of a cylinder-style combustion chamber with an inner diameter, 'D', of 90mm and a length of 1,000mm, and a primary air swirler and a fuel injection nozzle. The combustion chamber is lined with heat insulating material and the casing is cooled with water. There are four sections for secondary air on the sidewalls of the combustion chamber along the direction of flow, to simulate two-stage combustion. Secondary air inlets at a distance from the edge of the fuel injection nozzle of 3×'D' are used. The diameter of the secondary air inlets is 13mm, and altogether twelve inlets are positioned on the perimeter of each cross-section. The tested burner consists of a fuel injection nozzle and a primary air swirler. There are twelve injection inlets with a diameter of 1.5mm on the fuel injection nozzle with an injection angle, θ, of 90-degree. The primary air swirler has an inner diameter of 24.0mm, an outer diameter of 36.4mm, and twelve vanes with a swirl angle of 45-degree.

Table 1 Typical compositions derived from the oxygen-blown gasifiers

Fuel	Coal				Biomass	Heavy residue	Orimulsion ™
Gasifier type	Fixed	Entrained			Entrained	Entrained	Entrained
Fuel feed	Dry	Dry		Slurry			
Developer	BGL	Shell	HYCOL	Texaco		Texaco	CRIEPI
Composition							
CO	56.4%	65.2-67.8%	63.3%	40.9%	21.9-23.1%	51.7%	43.5%
H_2	25.6%	28.8-31.0%	26.3%	29.9%	12.5-22.4%	43.1%	42.2%
CH_4	6.6%	0.01-0.03%	0.3%	0.1%	2.2%	0.2%	0.4%
CO_2	2.8%	1.0- 2.8%	6.1%	9.5%	20.7-18.6%	3.2%	11.8%
H_2O	-(a)	(Dry base)	0.4%	12.3%	40.9-31.5%	(Dry base)	(Dry base)
NH_3	-(a)	100-600ppm	unknown	unknown	0- 200ppm	-(a)	unknown
H_2S+COS	20ppm	0.14- 1.1%	unknown	unknown	0.285-1.132%	1.6%	1.35%
Others(N_2)	8.6%	-(a)	8.6%	7.3%	1.800-1.048%	0.2%	0.75%
CO/H_2 mole ratio	2.2	2.1- 2.4	2.4	1.4	1.0-1.8	1.2	1.0
HHV[MJ/m^3]	13.0	12.2-12.5	11.5	9.0	5.2-6.6	12.1	11.0

(a): an omission

An air compressor supplies the air used for combustion. After the flow of the compressed air is adjusted to the required amount by the orifice meter and flow control valve, the air is heated to the prescribed temperature by an air heater, then blown into the combustor. Primary air is injected into the combustor through a primary air swirler positioned around the fuel injection nozzle, and secondary air is input through air holes in the sidewalls of the combustor.

CO and H_2, which are the main combustible components of the fuel, are mixed in the prescribed proportions and filled into a cylinder to be used. The combustible components are diluted with N_2 to create the required calorific value, heated to the prescribed temperature by an electric heater, and fed through the fuel injection nozzle. The small amounts of NH_3 and CH_4 are controlled by a thermal mass flow controller, and premixed with the fuel before reaching the fuel injection nozzle.

In this study, sample gases were extracted from the exit of the combustor through a water-cooled stainless steel probe and continuously introduced into an emissions console, which measured CO and CO_2 by infrared analysis, NOx by chemiluminescence analysis, O_2 by paramagnetic analysis, and hydrocarbons by flame ionization detector.

Reduction of NOx Emissions with Nitrogen Injection

To suppress thermal NOx production originating from nitrogen fixation, we designed the burner with nitrogen injection, based on combustion tests previously conducted using a small diffusion burner (Hasegawa, 1999b). Figure 2 presents an example of the test results using a small diffusion burner, under atmospheric pressure conditions, which indicates the influence of the primary-equivalence ratio, ϕ_p, on thermal-NOx emission characteristics in two-stage combustion for comparing three cases: 1) a fuel calorific value (HHV) of 12.7MJ/m^3, without nitrogen injection; 2) a fuel calorific value of 12.7MJ/m^3, where nitrogen is blended with the primary combustion air injected from the burner; 3) a fuel blended with nitrogen of the same quantity as case 2), or low-Btu fuel of 5.1MJ/m^3. In the tests, the fuel did not contain NH_3 and CH_4. A CO/H_2 molar ratio was set to 2.33, the combustor exit gas temperature, T_{ex}, was set to 1773K, and the fuel injection velocity was set at a constant value of 77m/s. The cross-sectional flow velocity of total air was 0.53m/s at the temperature of 273K basis in the case 2). From figure 2, we notice that nitrogen supply, which is blended with fuel or primary air, drastically decreases thermal-NOx emissions, and also NOx emissions decreases with rises in ϕ_p in the case of using the two-stage combustion. That is, thermal-NOx emissions decrease significantly by setting a fuel-rich condition when ϕ_p is 1.3 or higher in the case of nitrogen premixed with fuel, and by setting ϕ_p at 1.6 or higher in the case of nitrogen premixed with primary air.

With regard to fuel-NOx emissions on the other hand, figure 3 indicates the effects of nitrogen injection conditions on the conversion rate of NH_3 in the fuel to NOx, C.R., which is calculated from the following equation, in the same conditions with Fig. 2.

$$\text{C.R.}=\frac{([\text{NOx}]-[\text{NOx}_{th}])\times(\text{volume flow rate of exhaust})}{[\text{NH}_3]\times(\text{volume flow rate of fuel})} \quad \cdots\cdots\cdots\cdots\cdots (1)$$

Where $[NH_3]$ designates ammonia concentration in fuel.

To obtain the conversion rate of NH_3 to NOx, C.R., the concentration of thermal-NOx, '$[NOx_{th}]$', was first measured after stopping the supply of NH_3, then the concentration of total NOx, '[NOx]', was measured while NH_3 was supplied, and finally fuel-NOx was calculated by deducting the concentration of thermal-NOx from that of total NOx. In tests investigating fuel-NOx emissions, 1000ppm of NH_3 is contained in the medium-Btu fuel. In the case of a fuel blended with nitrogen, fuel was diluted, or fuel calorific value decreased to 5.1MJ/m^3 and NH_3 concentration in the fuel decreased to 400ppm. From figure 3, whether with or without nitrogen supplied, the staged combustion method effectively decreased the fuel-NOx emissions, or C.R. drastically decreased as the primary equivalence ratio, ϕ_p, become higher than 1.0, which is a stoichiometric condition, and shows the minimum value at the appropriate ϕ_p. Those optimum ϕ_p become lower when the medium-Btu fuel was blended with nitrogen, while the optimum ϕ_p was not influenced in the case of nitrogen blended with the primary combustion air injected from the burner, and C.R. showed a tendency to become a little higher than in the other two cases. Furthermore, under lean-lean combustion conditions with a lower ϕ_p than 1.0, in the case of nitrogen premixed with medium-Btu fuel, C.R. becomes higher than in the case of nitrogen premixed with the primary combustion air.

From the above, it was shown that the supply method of nitrogen premixed with medium-Btu fuel possibly decreases total emissions of thermal-NOx and fuel-NOx, but careful attention must be paid to the homogeneity of mixture of fuel and nitrogen, or thermal-NOx emissions will increase. In the case of nitrogen premixed with the primary combustion air, the power to compress nitrogen is lower than in the case of nitrogen premixed with fuel or the thermal efficiency of the plant increases, while total NOx emissions grow slightly higher. That is, it is necessary to blend nitrogen with medium-Btu fuel in the combustor, in which the lowest power to compress nitrogen is needed for nitrogen supply into the gas turbine, and not to collide the medium-Btu fuel with combustion air directly.

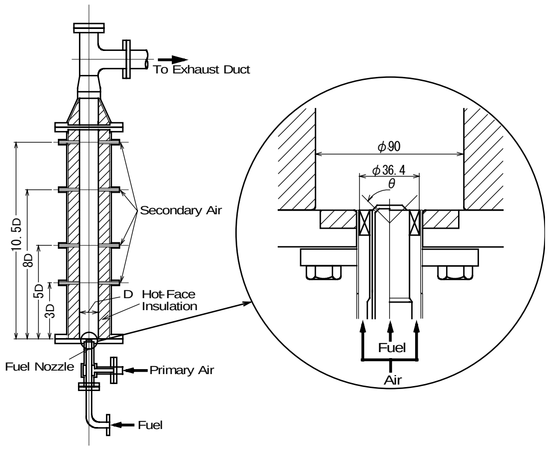


Fig.1 Combustor and diffusion burner of basic experimental device

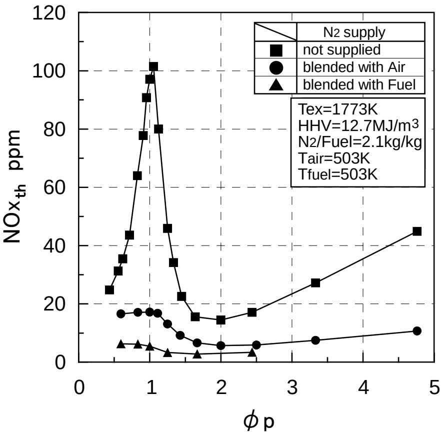


Fig.2 Effect of nitrogen injection on thermal-NOx emission characteristics in two-stage combustion, using a small diffusion burner

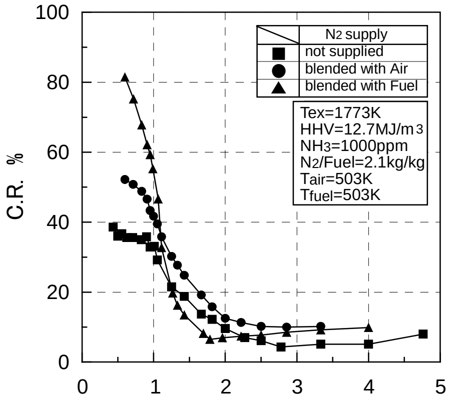


Fig.3 Effect of nitrogen injection on the conversion rate of ammonia to NOx in two-stage combustion, using a small diffusion burner

Effect of the Concentration of CH_4 in the Fuel

It is well known that the fuel-NOx production mechanisms of hydrocarbon fuels such as CH_4 and non-hydrocarbon fuels such as CO and H_2, due to reaction with nitrogenous compounds in the fuel, are different (Fenimore, 1976). Gasified coal fuel consists of CO and H_2 as main combustible components, but also contains thousands of ppm or a small percentage of CH_4. We therefore investigated the effects of CH_4 in the fuel on the production characteristics of fuel-NOx caused by NH_3 in the fuel.

Figure 4 shows the relationship between the primary equivalence ratio, ϕ_p, in two-stage combustion, and the conversion rate of NH_3 in the fuel to NOx with a concentration of CH_4 as a parameter. In the tests, the average temperature of combustor exit gas, T_{ex}, is set to 1773K and fuel calorific value is $11.4MJ/m^3$, for fuel containing 1000ppm of NH_3, CO and H_2 of 2.33 CO/H_2 molar ratio, and a small percentage of CH_4.

When the equivalence ratio of the primary zone, ϕ_p, is less than 1, the NH_3 to NOx conversion rate, C.R., increases in direct proportion to the concentration of CH_4 in the fuel; and when ϕ_p is more than 1, under fuel-rich conditions in the primary combustion zone and CH_4-free fuel, C.R. decreases as ϕ_p increases and shows a minimum at around 3 of ϕ_p. In the case of fuel containing CH_4, an optimum ϕ_p, at which C.R. drops to a minimum, is varied by adjusting the concentration of CH_4. That is, the optimum ϕ_p increases as the concentration of CH_4 decreases. For example, the optimum primary-equivalence ratio in the combustor have to be adjusted at 1.6-2.4, in the case where the simulated gasified fuel used in tests of the designed combustor contains around 3.0% CH_4.

The effects of the CH_4 concentration on the fuel-NOx produced by NH_3 in gasified coal fuel was studied using the elementary reaction kinetics (Hasegawa, 1999b). The nitrogen of NH_3 in the fuel has weaker bonding power than N_2. In the combustion process, NH_3 reacted with the OH, O, and H radicals and then easily decomposed into the intermediate NH_i by the following reactions.

$$NH_3 + OH(O, H) \Leftrightarrow NH_2 + H_2O(OH, H_2) \quad \cdots\cdots (2)$$

$$NH_i(i=1,2) + OH(H) \Leftrightarrow NH_{i-1} + H_2O(H_2) \quad \cdots\cdots (3)$$

When hydrocarbon is not contained in the fuel, NH_i is converted into N_2 by reacting with NO in the fuel-rich region. If fuel contains CH_4, HCN is produced by reactions 4 and 5 in the fuel-rich region and the HCN is oxidized to NO in the fuel-lean zone.

$$CH_i(i=1,2) + N_2 \Leftrightarrow HCN + NH_{i-1} \quad \cdots\cdots (4)$$

$$R\text{-}CH + NH_i \Leftrightarrow HCN + R\text{-}H_i, \quad (R\text{-}:\text{Alkyl group}) \quad \cdots\cdots (5)$$

With the rise in CH_4 concentration in gasified coal fuel, the HCN increases, and NOx emissions originated from HCN in the fuel-lean secondary combustion zone increase.

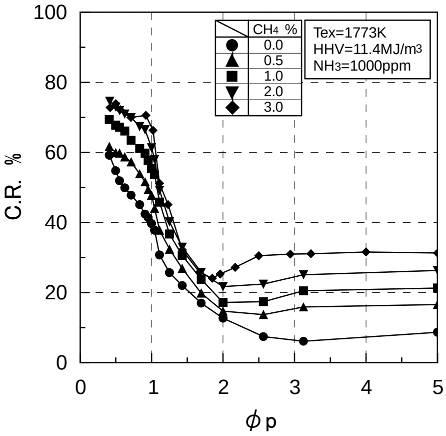


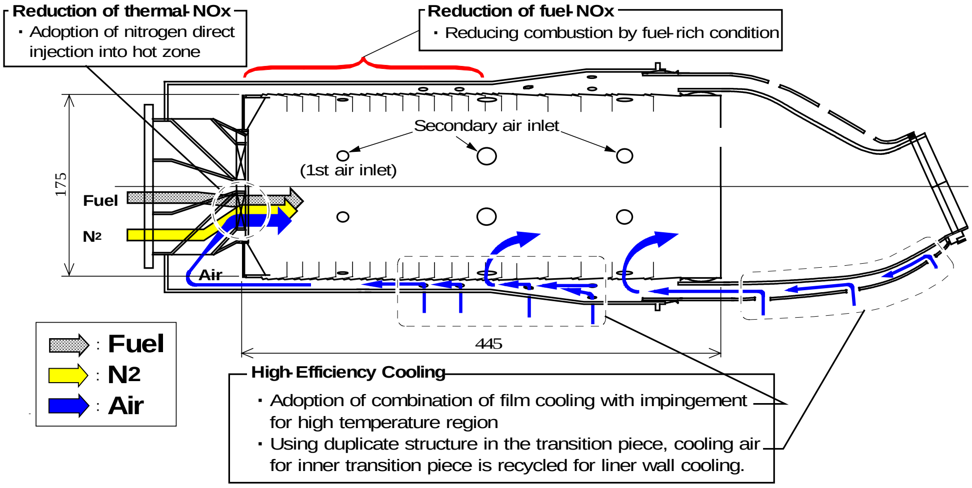
Fig.4 Effect of CH_4 concentration in fuel on the conversion rate of NH_3 in fuel to NOx in two-stage combustion, using a small diffusion burner

Characteristics of Tested Combustor

Figure 5 illustrates the configuration and its functions of a designed, medium-Btu fueled 1573K (1300℃)-class combustor based on the above considerations. The main design concepts for the tested combustor in the present study were to secure stable combustion of medium-Btu fuel with nitrogen injection in a wide range of turn-down operations, and low NOx combustion for reducing fuel-NOx and thermal-NOx emissions. In order to reduce thermal NOx emissions, we installed nitrogen injection nozzles in the burner. In order to reduce the fuel-NOx emissions, we adopted the two-stage combustion, in which a fuel-rich combustion was carried out in the primary zone maintaining the equivalence ratio of 0.56 at exit of the combustor. The overall length of the combustion liner is 445mm and the inside diameter is 175mm. And the designed combustor has another following characteristics.

(1) Tested burner

A tested burner used for this study was constructed for low thermal NOx emissions of medium-Btu fuel in the case where IGCC adopted a wet-type synthetic gas cleanup (Hasegawa, 1999a). We arranged the nitrogen injection intakes in the burner for low thermal-NOx emissions. Nitrogen injected directly into a combustor has the effect of decreasing power to compress nitrogen more than the each case of nitrogen premixed with fuel or air evenly. From system analysis, it was clarified that the thermal efficiency of the plant improved by 0.3 percent (absolute), compared with a case where nitrogen was premixed with gasified coal fuel before injection into the combustor. It is possible to control the mixing of fuel, air, and nitrogen positively by way of nitrogen being injected separately into the combustor. The fuel, the combustion air, and the nitrogen from the burner are separately injected into the combustor through a swirler, (which has a 30-degree swirl angle and a 15-degree introverted angle), to collide medium-Btu fuel with air in an atmosphere where nitrogen is superior in amount to both fuel and air.



Reduction of thermal-NOx
- Adoption of nitrogen direct injection into hot zone

Reduction of fuel-NOx
- Reducing combustion by fuel-rich condition

High-Efficiency Cooling
- Adoption of combination of film cooling with impingement for high temperature region
- Using duplicate structure in the transition piece, cooling air for inner transition piece is recycled for liner wall cooling.

Fig.5 Design concepts of a medium-Btu fueled gas turbine combustor

(2) Combustion liner

In order to decrease fuel-NOx emissions, we adopted fuel-rich combustion in the primary zone. This technique caused a decrease in flame temperature in the primary combustion zone, which produced thermal-NOx from nitrogen fixation. Thermal-NOx production near the burner was controlled just as in the case of fuel blended with nitrogen. Also, fuel-NOx emissions simultaneously decreased by using fuel-rich combustion, based on the results of combustion tests using a small diffusion burner. In the tests, we examined the effects of the primary equivalence ratio on the fuel-NOx emissions. Combustion liners had three sections for secondary air inlets along the direction of flow and altogether eight inlets were positioned on the perimeter of each cross-section. The combustion liner was divided four zones, axial distance of each was equal, and the equivalence ratios in the primary combustion zone from the edge of the burner to second air inlet section, ϕ_p, were set to 1.35 (type-1 combustor) and 1.60 (type-1 combustor) of each combustion liner, respectively. Type-2 combustor had no first inlets of the secondary air, so that ϕ_p was set to around 1.6, in particular.

The designed combustor was given another nitrogen injection function, in which nitrogen was by-passed to premix with the air derived from the compressor, and a stable flame can be maintained in a wide range of turn-down operations. Also, because the nitrogen dilution in the fuel-rich region affected the deoxidization characteristics of NH_3, the nitrogen bypassing technique was expected to improve fuel-NOx reduction in the cases of higher concentration of NH_3. For cooling of the overall combustion liner wall, the film-cooling method was used. For the secondary-air inlet section where temperatures were expected to be especially high, the dual-cooling structure was employed, in which the cooling air was impinged from the air flow guide sleeve to the combustion liner.

(3) Transition piece

In order to compensate for a declined cooling-air ratio associated with a surplus nitrogen injection into a gas turbine combustor, the tested combustor is equipped with a dual-wall structure transition piece so that the cooling air in the transition piece can be recycled to cool down the combustor liner wall. The cooling air, flowing into the transition piece from the exterior wall, cools the interior wall through an impingement and convection method, and moves to the combustor liner on the upstream side.

Table 2 shows the typical compositions of the supplied fuel and the expected actual gasified fuel. As for tests, the higher heating value (HHV) of the supplied fuel was set at 8.8MJ/m^3, and a $(CO+CH_4)/H_2$ molar ratio at 1.2. The flow rate of surplus nitrogen produced from the air-separation unit was 0.9～1.2 times the fuel flow in the actual process. Since the density of the supplied fuel was higher than that of the gasified coal fuel, and since the temperature of supplied nitrogen was lower in test conditions than in the actual operations, we also investigated combustor performance in the case of a 0.7kg/kg N_2/fuel ratio, where the firing temperature of the burner exit corresponded actual operation. Rated load conditions in the combustion tests are summarized in table 3. The combustor-exit gas temperature was 1673K and the combustion intensity in the combustor at the design point was $3.9\times10^2 W/(m^3{\cdot}Pa)$.

Table 2 Typical compositions of supplied fuel and actual gasified fuel

		supplied fuel	actual gasified fuel
Composition	CO	31.4 %	40.9 %
	H_2	28.6 %	29.9 %
	CH_4	3.0 %	0.1 %
	CO_2	32.0 %	9.5 %
	H_2O	-	12.3 %
	N_2	5.0 %	7.3 %
	NH_3	2100 ppm	0～3000 ppm
HHV		8.8 MJ/m^3	9.0 MJ/m^3
LHV		8.1 MJ/m^3	8.2 MJ/m^3

Table 3 Standard test conditions

T_{air}	:	603K
T_{fuel}	:	583K
T_{N_2}	:	333K
N_2/Fuel	:	0.70 kg/kg
T_{ex}	:	1673 K
ϕ_{ex}	:	0.56
P	:	1.2 MPa
Ic	:	$3.9\times10^2 W/(m^3{\cdot}Pa)$
Ur	:	5.6 m/s
$\Delta P/q$	:	1.64×10^2

TEST FACILITIES AND TEST METHOD

Test Facilities

The schematic diagram of the test facilities is shown in figure 6. The raw fuel obtained by mixing CO_2 and steam with gaseous propane was decomposed to CO and H_2 inside the fuel reforming device. A hydrogen separation membrane was used to adjust the CO/H_2 molar ratio. N_2 was added to adjust the fuel calorific value to the prescribed calorie, and then coal-derived simulated gases were produced.

This facility had another nitrogen supply line, by which nitrogen was directly injected into the combustor. Air provided to the combustor was pressurized to 2.0MPa by using a four-stage centrifugal compressor. Both fuel and air were supplied to the gas turbine combustor after being heated separately with a preheater to the prescribed temperature.

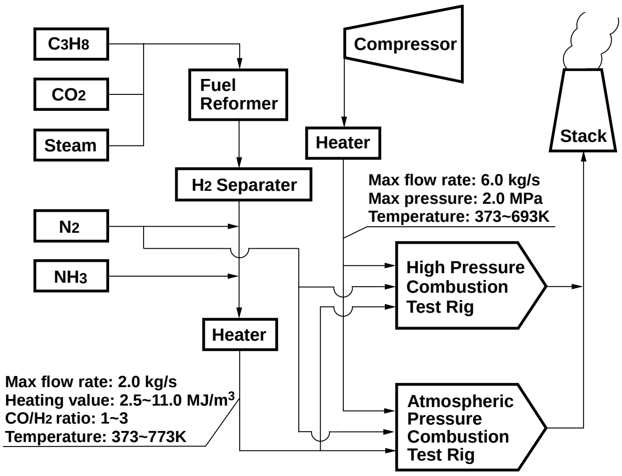


Fig.6 Schematic diagram of experimental facility

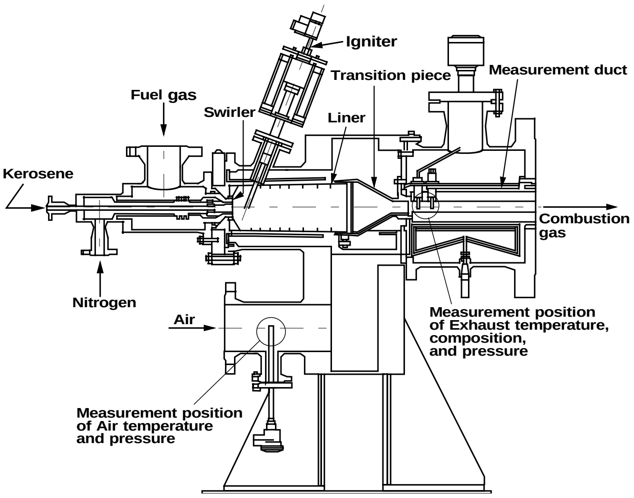


Fig.7 Combustion test rig

The combustion test facility had two test rigs, each of which was capable of performing full-scale atmospheric pressure combustion tests of a single-can for a "several"-hundreds MW-class, multican-type combustor as well as half-scale high-pressure combustion tests, or full-scale high-pressure tests for around a 100MW-class, multican-type combustor. Figure 7 shows a cross-sectional view of the combustor test rig under pressurized conditions. After passing through the transition piece, the exhaust gas from the combustor was introduced into the measuring section where gas components and temperatures were measured. The components of the combustion gases were analyzed by an automatic gas analyzer. After that, the gas temperature was lowered through a quenching pot, using a water spray injection system.

Combustion tests were conducted on a full-scale, single-can combustor for a 100MW-class gas turbine under high-pressurized conditions.

Measurement System

Sample gases were taken from the exit of the combustor through water-cooled stainless steel probes located on the centerline of a height-wise cross section of the measuring duct. The sample lines of exhaust gases were thermally insulated with heat tape to maintain the sampling system above the dew point of the exhaust gas. The gas samples taken were from an average of sampling points on the center-line of the measuring duct and continuously introduced into an emission console which measured CO, CO_2, NO, NOx, O_2, and hydrocarbons by the same methods as the test device for basic studies using the small diffusion burner. The medium-Btu simulated fuel were sampled from the fuel gas supply line at the inlet of combustor, and CO, H_2, CH_4, H_2O, CO_2 and N_2 were determined by gas chromatography. Heating values of the simulated gaseous fuel were monitored by a calorimeter and calculated from analytical data of gas components obtained from gas chromatography.

The temperatures of the combustor liner walls were measured by 40 sheathed type-K thermocouples with a diameter of 1mm attached to the liner wall with a stainless foil welding. The temperature distributions of the combustor exit gas were measured with an array of three pyrometers, each of which consisted of five type-R thermocouples.

TEST RESULTS AND DISCUSSION

NOx Emission Characteristics

We carried out researches into the effects of the primary equivalence ratio, combustor exit temperature, sectional flow velocity, nitrogen bypassing method, and combustion pressure on NOx emission characteristics.

Figure 8 shows the relationship between the average temperature of combustor-exit gas, T_{ex}, and the conversion rates of fuel-NOx originating from NH_3 in the fuel and thermal-NOx emissions, and when: air flow rate was set and maintained at 2.1kg/s, the nitrogen flow rate of N_2/fuel was 0.7kg/kg, the pressure inside a combustor was at a slightly less 1.2MPa than that of the practical rated operation, and NH_3 and CH_4 concentration in the fuel were 2100ppm and 3.0% respectively. In the tests, all of the nitrogen was injected from the burner. As the average temperature of combustor-exit gas, which was equivalent to the gas turbine load condition, was varied, the fuel flow rate was changed to maintain the airflow rate at a constant value of 2.1kg/s and the flow ratio of nitrogen injection over fuel (N_2/Fuel) at constant. In the case of type-1 combustor (ϕ_p=1.35), as the average temperature of combustor-exit gas was raised to 1173K, which corresponds to 25% of the gas turbine load, or higher, the conversion rate of NH_3 in the fuel to NOx, C.R., showed a tendency to decrease gradually, while thermal-NOx emissions were reduced as low as 6ppm (corrected at 16% O_2). In the case of type-2 combustor (ϕ_p=1.60), the conversion rate was declined around 10% absolutely compared with the case of type-1 combustor. On the other hand, thermal-NOx emissions were raised slightly compared with the case of type-1 combustor. At each case, nitrogen injection decreased the local high-temperature regions and thermal-NOx emissions decreased. And also, with the rise in the primary equivalence ratio, ϕ_p, from the 1.35 (type-1 combustor) to 1.60 (type-2), C.R. was decreased to around 40% in the case where the fuel contained NH_3 around 2100ppm. If the medium-Btu fuel was mixed with the nitrogen produced from air separation unit, the fuel calorific value was decreased to around 5MJ/m^3 and NH_3 concentration to around 1100ppm, they were same levels in the cases of low calorific fuels (Hasegawa, 1998a). That is to say, in case of medium-Btu fuel, the conversion rates could be decreased as low as that of low-Btu fuel. Furthermore, CO emissions were reduced as low as 20ppm at any gas turbine load, or combustion efficiency was kept higher than 99.9%.

Figure 9 shows the effect of the cross-sectional velocity of air, Ur, on the NOx emission characteristics at the rated temperature of 1673K, in the case of type-1 combustor. In the tests, the nitrogen flow rate of N_2/fuel was set at 0.7kg/kg, the pressure inside a combustor was at 1.2MPa, and NH_3 and CH_4 concentration in the fuel were 2100ppm and 3.0% respectively, just as in the case of Fig.8. All of the nitrogen was injected from the burner. With the rise in Ur, thermal-NOx emissions have hardly changed, while the conversion rate of NH_3 to fuel-NOx showed a tendency to increase. That is, the residence time of the combustion gas in the combustor declined with the rise in Ur, but thermal-NOx emissions was not changed and maintained low level of 10ppm(corrected at 16% O_2) or below. On the other hand, reduction of NH_3 into N_2 in the primary combustion zone was declined and fuel-NOx emissions increased. If the residence time in the reduction combustion zone could be increased, the fuel-NOx emissions will be restrained more, while the thermal NOx emissions maintain 10ppm or below.

Nitrogen supply positions affected temperature distribution, thermal-NOx, and fuel-NOx production in the combustor. We observed NOx emission characteristics when a part of the nitrogen was injected into the combustor through the burner and the rest was bypassed to premix with the combustion air, which was injected from the burner-air nozzles and secondary combustion air holes. Figure 10 shows the relationship between the ratio of bypassing nitrogen flow rate over total nitrogen supply, N_2(BY)/N_2total, and both the conversion rate of NH_3 to NOx, C.R., and thermal-NOx emissions, in the each combustor. In the tests, the average temperature of combustor-exit gas was maintained at 1673K and another conditions were just as same as in the case of Fig.8.

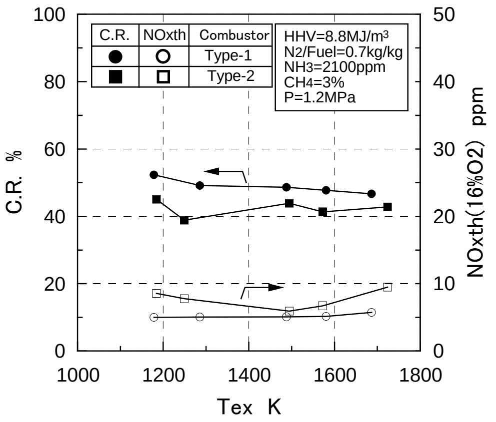


Fig.8 Effect of combustor-exit gas temperature on NOx emission characteristics in the case of nitrogen injection

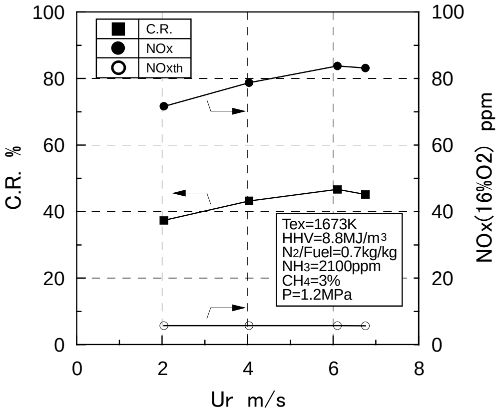


Fig.9 Effect of sectional flow velocity of air in the combustor on NOx emission characteristics in the case of nitrogen injection

At each combustor, the conversion rate slightly decreased with the rise in the N_2(BY)/N_2total ratio, while thermal-NOx emissions gradually increased. Since the conversion rate declined with the rise in NH_3 concentration (Hasegawa, 1999b), the nitrogen bypassing method is expected to be effective in the case of gasified fuel contained higher concentrations of NH_3.

In the combustor tests, the maximum pressure was set to a slightly less of 1.2MPa than that of the practical operation at the equivalent, rated load conditions for restrictions of the fuel supply rate. So, we examined the effects of pressure inside a combustor on the NOx emission characteristics. Figure 11 shows the relationship between the pressure inside the combustor, and both the conversion rate and thermal-NOx emissions in the case of type-1 combustor. In the tests, all of the nitrogen was injected from the burner or bypassed into the combustion air, and the pressure inside a combustor was changed to maintain the residence time of the combustion gas in the combustor at constant. In the case of nitrogen injected directly from the burner, the conversion rate of NH_3 to NOx hardly changed with the rise in pressure from 0.4MPa to 1.2MPa, and the thermal-NOx emissions slightly increased. On the other hand, when the nitrogen bypassed into the combustion air, the flame temperature near the burner became higher than the case of nitrogen direct injection, thermal-NOx emissions increased twice that in the case of the nitrogen direct injection, while the conversion rate showed a tendency to decrease. The pressure indexes, P_{index}, which indicates the inclination of the logarithmic values of the thermal-NOx emissions against the pressure as shown by the following equation, were around 0.2～0.3 in each case.

$$\frac{[NOx]_2}{[NOx]_1} = \left(\frac{P_2}{P_1}\right)^{P_{index}} \quad \cdots\cdots \quad (6)$$

Where the subscripts 1 and 2 designate the emissions and the pressures in the combustor at the respective conditions.

As an example of the effect that pressure exerts on the level of the thermal-NOx emissions from the nitrogen fixation in the gas turbine combustor using hydrocarbon fuels, it is known that the pressure to the power of 1.5 is nearly proportional to the speed at which the Zel'dovich NO is produced (i.e. $d[NO]/dt \propto P^{1.5}$) and that the NOx emission mole fraction varies with pressure to the 0.5 power. However, in the case of the medium-Btu fueled combustor with nitrogen injection, it is found that influence of the pressure on the thermal-NOx emissions showed the different tendency from that of the hydrocarbon fuel and lower dependence of the thermal-NOx emissions on pressure. That is because, the volume of the medium-Btu fuel was larger than that of hydrocarbon fuels, and so mixing characteristics was improved. Furthermore, since the fuel-rich combustion where produced Prompt NO was employed in the first combustion zone, thermal-NOx emissions showed a different tendency from the case of hydrocarbon fuels. So the thermal-NOx emissions showed the weaker dependence on pressure.

With respect of the difference in effects of pressure on the conversion rate, we estimated as follows:

NH was produced by NH_3 decomposition reaction 2, 3, and by HCN formation reaction 4.

$$NH_3 + OH(O, H) \Leftrightarrow NH_2 + H_2O(OH, H_2) \quad \cdots\cdots \quad (2)$$

$$NH_i(i=1,2) + OH(H) \Leftrightarrow NH_{i-1} + H_2O(H_2) \quad \cdots\cdots \quad (3)$$

$$CH_i(i=1,2) + N_2 \Leftrightarrow HCN + NH_{i-1} \quad \cdots\cdots \quad (4)$$

Some HCN is oxidized into NO by reactions 7 and 8, and the rest is decomposed into N radical by the reaction 9. NH radical is oxidized into the NO by reactions 10, 11, and 12.

$$HCN+OH \Leftrightarrow CN+H_2O \quad \cdots\cdots \quad (7)$$

$$CN+O_2 \Leftrightarrow CO+NO \quad \cdots\cdots \quad (8)$$

$$CN+O \Leftrightarrow CO+N \quad \cdots\cdots \quad (9)$$

$$NH+OH \Leftrightarrow N+H_2O \quad \cdots\cdots \quad (10)$$

$$N+O_2 \Leftrightarrow NO+O \quad \cdots\cdots \quad (11)$$

$$N+OH \Leftrightarrow NO+H \quad \cdots\cdots \quad (12)$$

On the other hand, some NH radical produced by the reactions 2 and 3, 4 are reacted with Zel'dovich NO, Prompt NO and fuel-N oxidized NO, which produced by above reactions, and decomposed into N_2 by the reaction 13.

$$NO+NH \Leftrightarrow N_2+OH \quad \cdots\cdots \quad (13)$$

That is, it is surmised that the pressure inclination affected the thermal-NO and fuel-NO productions, and alternative decomposition reaction of intermediate NH radical with NO increased, so the conversion rate of NH_3 in fuel declined.

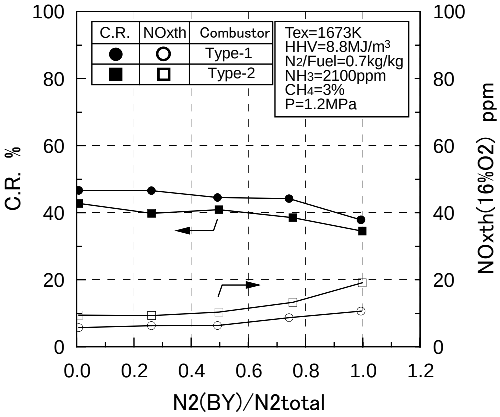


Fig.10 Effect of bypassing nitrogen flow rate, which premixed with combustion air on NOx emission characteristics

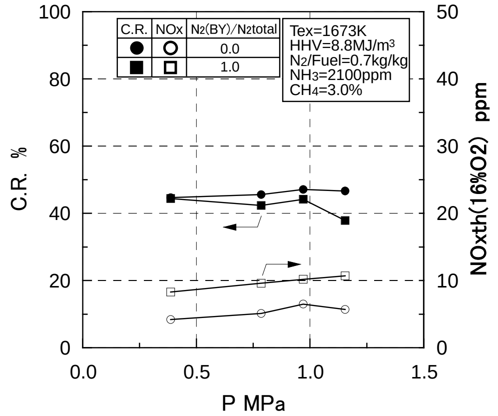


Fig.11 Effect of pressure inside a combustor on NOx emission characteristics

In the case of nitrogen direct injection, since the thermal-NOx production was as low as 5ppm, decomposition of fuel-NOx was not expected, or C.R. was hardly influenced by pressure increase.

CONCLUSIONS

With the preliminary combustion tests using a small diffusion burner and a combustor newly designed with a suitable nitrogen injection nozzle, we examined the effects of two-stage combustion and nitrogen injection on the NOx emission characteristics. Results are summarized as follows:

(1) From the combustion tests of the medium-Btu fueled combustor, we clarified that the two-stage combustion with nitrogen direct injection into the combustor effective in reductions of fuel-NOx and thermal-NOx emissions.

(2) In the tested combustor, the conversion rate of NH_3 to NOx could be decreased to 40% and thermal-NOx emissions to 10ppm(corrected at 16% O_2) or less under gas turbine operational conditions of 25% load or higher for IGCC in the case where the gasified fuel contained 3% CH_4 and 2100ppm NH_3.

(3) Elongation of the residence time of the combustion gas could decrease the fuel-NOx emissions more, while thermal-NOx emissions was hardly changed.

(4) Nitrogen bypassing way decreases the conversion rate of NH_3 in the fuel, while thermal-NOx emissions increases. That is, in the case of higher NH_3 concentration, bypassing methods of all of nitrogen or some part of nitrogen are expected to be effective for reducing total NOx emissions.

ACKNOWLEDGEMENTS

The authors wish to express their appreciation to the many people who have contributed to this investigation. In testing this combustor, we received helpful support from Mr. Kawashima,K. and Kousaka,Y. (Central Research Institute of Electric Power Industry), Mr. Sezaki,H., Suzuki,M., and Terada,K. (Techno Service Corp.).

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