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Study on Freeboard Properties to Maintain Low N_2O Emissions from Sewage Sludge in a Fluidized Bed Combustor

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A new type of incineration plant, which was equipped with a pressurized bubbling fluidized bed combustor coupled with a turbocharger, was proposed. N_2O emissions in the proposed combustor of the demonstration plant were less than half of those observed in an atmospheric plant. In this study, the freeboard properties in the combustor were investigated to clarify the detailed reason for the low N_2O emissions under pressurized conditions. First, the effect of a wide range of operating pressures on N_2O emissions in a laboratory-scale pressurized fluidized bed combustor, which can control the freeboard temperature independently using an electric heater, was examined experimentally. Second, the effect of the operating pressure on the freeboard temperature profiles was estimated theoretically by a computational analysis. As a result, N_2O emissions were obviously decreased with an increasing freeboard temperature even when the operating pressure changed substantially. Thus, the freeboard temperature is an important factor to reduce N_2O emissions in the elevated pressure conditions as well as in the atmospheric conditions. Additionally, it was verified that the attainment point of the highest temperature shifted toward the bottom of the freeboard with increasing pressure by the computational analysis. This finding corresponded with the freeboard temperature profiles in the demonstration-scale combustor.

Introduction

The annual production of sewage sludge in Japan increased to 2.23 million tons [dry basis (db)] in 2005, and more than 70% of this sewage sludge is incinerated.[1−3] Because the nitrogen content of this sludge is considerably higher than that of other fuels, such as coal and wood, NO_x and N_2O emissions from the sludge are expected to be high. In particular, the reduction of N_2O emissions is required from the viewpoint of the global warming issue, because the global warming potential of N_2O is 310 times as high as that of CO_2. In previous studies, the effect of the operating pressure on N_2O emissions in pressurized fluidized bed combustion of coal has been studied.[4,5] Mallet et al. have clarified that the amount of N_2O formed from volatile N was larger than that from char N in low-rank coal.[6] N_2O emissions were found to decrease with an increasing O/N molar ratio in the coal because the amount of NH_3 released during devolatilization increases with an increasing O/N ratio.[7,8] In addition, N_2O reduction by coal char particles has been investigated in a packed bed reactor in the presence of N_2O/N_2 mixtures.[9,10] Our previous study has been reported; NO_x and N_2O emissions in the demonstration plant in a new incineration system, which combines a pressurized fluidized bed combustor (PFBC, design pressure = 0.3 MPa) and a turbocharger, were less than half of those observed in a conventional atmospheric plant.[11] The dependency of N_2O emissions from the sludge upon the freeboard temperature in an atmospheric fluidized bed combustor has been reported.[12] Figure 1 shows the comparison of the temperature distributions in the atmospheric combustor to the pressurized combustor by the demonstration-scale plant.[13] The temperature rises sharply in the lower part of the freeboard under pressurized conditions. It was thought that the pressurization may also have suppressed N_2O emissions because N_2O was decomposed at the locally formed high-temperature region in the freeboard. However, it has not yet been clarified why the difference of the freeboard temperature distribution by the operating

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pressure occurs. The objective of our present study is to clarify the reason for the low N_2O emissions under elevated pressure conditions by considering the freeboard properties in the combustor. The relationship of N_2O emissions and the freeboard temperature was elucidated in the wide range of operating pressure conditions using a laboratory-scale PFBC. Additionally, the effect of the operating pressure on the freeboard temperature distribution was estimated theoretically by computational analysis.

Experimental Section

Fundamental Combustion Test Using a Laboratory-Scale PFBC. The laboratory-scale PFBC system consists of a sludge pump, a bubbling fluidized bed combustor, a ceramic filter, a pressure vessel, and a compressor; the schematic diagram is illustrated in Figure 2.[14] The pressure vessel is made of stainless steel and is 1200 mm in internal diameter and 3200 mm in height. The fluidized bed combustor is made of stainless steel and is 80 mm in internal diameter and 1300 mm in height. The temperatures were measured by thermocouples located at heights above the distributor: 50 and 300 mm in the bed and 800 and 1200 mm in the freeboard. Silica sand with a mean particle size of 240 μm was employed as the bed material, and the static bed height was about 300 mm. The properties of the fuel used in the experiments are listed in Table 1. This sludge was received in sewage works near Tsukuba.

The combustor was installed into the pressure vessel, and compressed air was supplied to the vessel. The inside pressure was controlled by adjusting the inlet air flow rate with a mass flow controller. The flow rate of the combustion air supplied through the distributor was measured at the bottom of the combustor by a mass flow meter. The sand bed and the freeboard temperatures were controlled independently by the electrical heater. Dewatered sludge was fed into the fluidized bed continuously by a high-pressure sludge pump through an injection nozzle located at the top of the combustor. The sludge feeding was initiated when the bed temperature exceeded a predetermined temperature. The flue gas was exhausted into the atmosphere through a letdown valve after passing through a ceramic filter to remove dust.[15,16]

The fuel was supplied to the combustor to keep the O_2 concentration of 4−8% in the flue gas. The freeboard temperature was maintained in the range of 1120−1300 K, and the absolute furnace pressure was adjusted to 0.2, 0.3, and 0.6 MPa. The superficial air velocity was kept constant at 0.15 m/s under

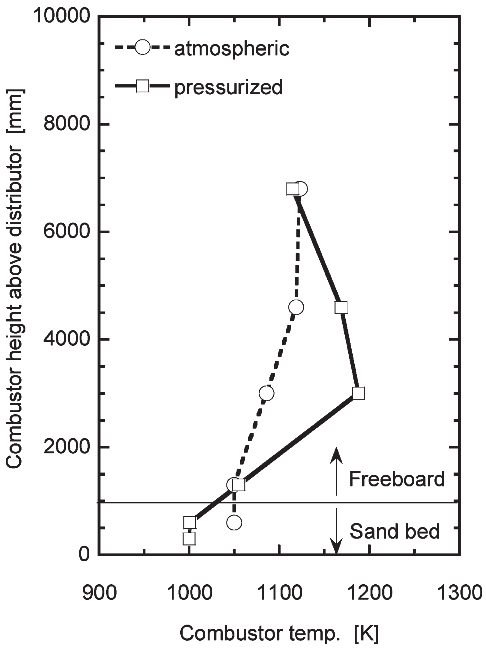

Figure 1. Comparison of temperature distributions observed in combustors operated under atmospheric and pressurized conditions.

Table 1. Properties of the Sewage Sludge Used for Fundamental Combustion Tests

proximate analysis (wet, wt %)	
moisture	78.0
volatile matter (VM)	13.9
fixed carbon (FC)	1.8
ash	6.3
ultimate analysis (dry, wt %)	
C	29.8
H	4.0
N	5.0
S	1.1
O	31.4
higher heating value (MJ/kg, db)	17.1

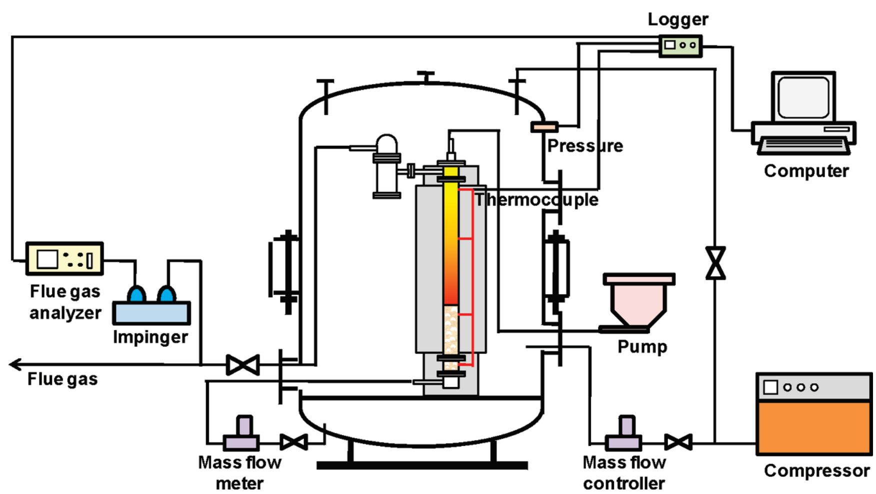

Figure 2. Schematic diagram of a laboratory-scale PFBC system.

Table 2. Properties of the Fuel Used for Calculations

	dewatered sludge	heavy oil
proximate analysis (wet, wt %)		
moisture	86.6	
VM	9.89	
FC	1.49	
ash	2.02	
ultimate analysis (dry, wt %)		
C	43.6	86.5
H	6.97	12.6
N	7.27	0
S	0.82	0.9
O	26.2	0
higher heating value (MJ/kg, db)	20.2	45.2

Table 3. Estimated Gas Compositions at the Inlet of the Freeboard

gas	mole fraction
H_2	0.0039
CO	0.0097
CH_4	0.0117
CO_2	0.0513
H_2O	0.3199
O_2	0.0585
N_2	0.5450

all operating pressures. A portion of the flue gas was bypassed to impingers in series to remove moisture, and then O_2, CO, CO_2, and NO_x concentrations in the flue gas were analyzed with continuous gas analyzers (IR400 Yokogawa Electric Corp., Tokyo, Japan). The N_2O concentration was measured with a gas chromatograph (G2891A Hewlett-Packard, Palo Alto, CA) at 3 min intervals.

Computational Analysis of Homogeneous Gas-Phase Combustion in the Demonstration-Scale Combustor. A software package for the simulation of homogeneous gas-phase combustion reactions, CHEMKIN III, was used to analyze the freeboard temperature profile process in the demonstration-scale combustor under the elevated pressure conditions. The freeboard in the combustor was simplified as a one-dimensional homogeneous gas-phase chemical reactor to simulate combustion in the demonstration-scale combustor. The "PLUG", a calculation code of the plug flow reactor, was employed in this calculation. All of the elemental combustion reaction equations were selected from the list shown on the GRI-Mech website,[17] where 49 chemical species and 277 elementary reactions were listed. Thermochemical constant and transport coefficient of each element were calculated using the CHEMKIN subroutine. The calculation conditions were as follows:[11] The feeding rates of dewatered sludge and the auxiliary fuel (heavy oil) were fixed at 180 and 20 kg/h, respectively. Their properties of the fuel used in the calculations are listed in Table 2. This sludge was used by the demonstion-scale combustor in Oshamanbe Cho in Hokkaido, Japan. The air ratio was selected as 1.5. The heat loss was omitted from the calculation because the combustor was assumed to operate under adiabatic conditions. The operating absolute pressure conditions were 0.1 and 0.3 MPa. The initial superficial gas velocity was 1.5 m/s. The height of the freeboard was 8200 mm.

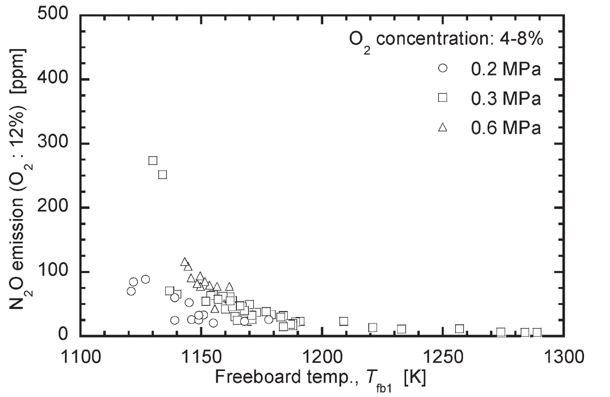

Figure 3. Relationship between N_2O emissions in the flue gas and the freeboard temperature. T_{fb1} is the temperature measured by the thermocouple located at a height of 800 mm above the distributor in the combustor.

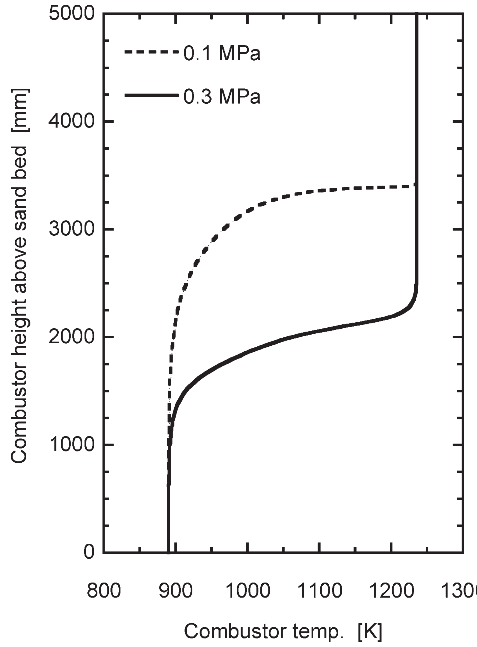

Figure 4. Effect of the pressure on the temperature distribution in the freeboard by calculation.

In addition, the gas compositions at the inlet of the freeboard were required for this calculation. However, determining the gas compositions at the inlet of the freeboard is difficult. Therefore, the major gas compositions were estimated by the assumption that the drying and pyrolysis of the dewatered sludge, as well as the combustion of heavy oil, were completed in the sand bed. From these assumptions, the major gas compositions (H_2, CO, CH_4, CO_2, H_2O, O_2, and N_2) were estimated on the basis of the mass and energy balance of the fuel (Table 3). Gases, such as SO_2, were omitted because their amounts of concentrations were negligible. Thus, the calculation started from the bottom of the freeboard and proceeded toward the exit of the combustor.

Results and Discussion

Flue Gas Characteristics in the Fundamental Combustion Test. Figure 3 shows the relationship between the N_2O

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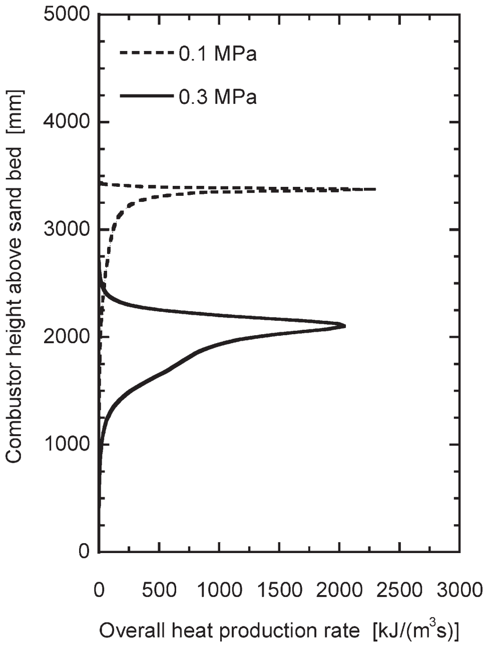

Figure 5. Effect of the pressure on overall heat production rates in the freeboard by calculation.

emissions and the freeboard temperature. Increasing the operating pressure resulted in a slight increase in N_2O emissions. However, the emissions are obviously decreased with an increasing freeboard temperature.[4,5] These data show that the freeboard temperature can control N_2O emissions in the pressurized combustor as well as the atmospheric combustor.[12] The finding suggests that the N_2O concentration is decreased substantially because of N_2O destruction reactions at the locally formed high-temperature zone of the freeboard under the pressurized conditions in the demonstration-scale combustor; thus, final N_2O emissions can be reduced.

Computational Analysis of Homogeneous Gas-Phase Combustion in the Demonstration-Scale Combustor. Figure 4 shows the calculated temperature profiles in the freeboard of the combustor at different operating pressures. A sharp temperature increase in the lower part of the freeboard occurs with an increasing pressure, and the position of the highest temperature moves toward the sand bed surface. Figure 5 shows a comparison of the overall heat production rate, which is calculated by the total amount of the heat production rate of each elementary reaction. The peak of the overall heat production rate corresponds to the sharp temperature rise region. It is thought that the chemical reaction rates of particular elementary reactions are increased at elevated pressure because the pressurization effectively increases the molar densities of the chemical species. The typical reactions are shown as follows:

$$HCO + O_2 \leftrightarrow HO_2 + CO \qquad (1)$$

$$OH + CH_2O \leftrightarrow HCO + H_2O \qquad (2)$$

$$2HO_2 \leftrightarrow O_2 + H_2O_2 \qquad (3)$$

$$OH + HO_2 \leftrightarrow O_2 + H_2O \qquad (4)$$

$$OH + CO \leftrightarrow H + CO_2 \qquad (5)$$

Consequently, the temperature in the freeboard might rise sharply in the lower part with an increase of the operating pressure. A more comprehensive model may be required to explain the typical temperature distribution observed under pressurized conditions. Nevertheless, the main cause of low N_2O emissions in the demonstration-scale combustor was verified here.

Conclusions

The fundamental combustion tests using the laboratory-scale PFBC and the computational analysis were carried out to clarify the reason for the low N_2O emissions in the flue gas under the elevated pressure conditions in the fluidized bed combustor of sewage sludge. From the fundamental combustion tests, N_2O emissions were only weakly dependent upon the operating pressure but were strongly dependent upon the freeboard temperature. Therefore, N_2O emissions can be arranged by the freeboard temperature, even if the operating pressure changes. Additionally, the change in the freeboard temperature profile by operating pressure in the demonstration-scale combustor was verified qualitatively by the computational analysis based on the elemental homogeneous reaction that occurred in the combustor.

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