

## A Study of Nitrous Oxide Decomposition over Calcium Oxide in Combustion Condition

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**Abstract**—A study of N<sub>2</sub>O decomposition reaction over a bed of CaO particles in a fixed bed reactor has been conducted. Effects of parameters such as concentration of inlet N<sub>2</sub>O, reaction temperature and effects of CO, CO<sub>2</sub>, O<sub>2</sub>, and NO gas presented in the combustion gas environment have been investigated. The experiment showed that the N<sub>2</sub>O decomposition reaction was accelerated by the increase of reaction temperature, and the existence of CO, while the reaction was hindered by the existence of CO<sub>2</sub>, NO. O<sub>2</sub> also affected the N<sub>2</sub>O decomposition. Heterogeneous gas-solid reaction kinetics were proposed for the reaction conditions and compared with homogeneous reaction kinetics.

Key words : Nitrous Oxide Decomposition, Calcium Oxide, Kinetics

### INTRODUCTION

The emission of N<sub>2</sub>O has been recognized as one of the major causes of global warming and harmful effect on the ozone layer in the upper atmosphere. The N<sub>2</sub>O, which is emitted to the environment mostly during natural processes, has been reported to have over 200 times stronger greenhouse effect than CO<sub>2</sub>. Major source of N<sub>2</sub>O generation is the oxidation of N<sub>2</sub> component by bacteria in the soil, destruction of the forests and land clearing, and excessive utilization of nitrogenous fertilizers. Also, the incineration of wastes and the power generation using fossil fuel can be another source [de Soete, 1994]. Around 10% of N<sub>2</sub>O release is reported to be contributed by coal combustion process.

The circulating fluidized bed (CFB) boilers have been welcomed in utility and waste incineration business for their excellent combustion and boiler efficiency. However, the fluidized bed combustor is reported to release significantly higher amount of N<sub>2</sub>O than other types of coal burning combustors [Åmand and Andersson, 1989]. There were a few suggestions for reduction or destruction of N<sub>2</sub>O leaving CFB combustor such as by increasing the combustor temperature [Suzuki et al., 1996], but the method is difficult to apply since keeping the inside combustor temperature above 950 °C may cause damage in the boiler parts and increase the opportunities for clinker formation.

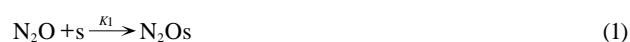
For the CFB combustors limestone (CaCO<sub>3</sub>) is usually injected with feed coal for sulfur removal and the limestone also decompose N<sub>2</sub>O over unused CaO. Thus the emission is suppressed by the existence of CaO [Chang et al., 2000; Johnson and Jensen, 1994; Peter et al., 1993]. There are gas-solid reactions on the surface of CaO with N<sub>2</sub>O, and the reaction order is suggested to be first order [Shimizu et al., 1993]. In this study the decomposition of N<sub>2</sub>O over CaO with various combustion gas components, which simulates the gas composition of lower parts of CFB combustor, was conducted. The effects of reacting temperature and content of CO<sub>2</sub>, CO, NO, and O<sub>2</sub> on N<sub>2</sub>O decomposition were analyzed, and a kinetic

study considering the catalytic effect of CaO was conducted [Klvana et al., 2002; Panczyk and Ryzinski, 2004].

The kinetic approach of N<sub>2</sub>O decomposition over calcium oxide has been suggested by many authors on either case of fluidized bed combustion [Johnsson et al., 1997; Satsuma et al., 2000; Shimizu et al., 1993] or fixed bed combustion [Satsuma et al., 2000]. However due to the complex nature of N<sub>2</sub>O decomposition mechanism, most efforts were concentrated on the generalization on overall kinetics. In order to understand the fundamental role of each constituent in the combustion environment, this work concentrated on the role of major combustion products on N<sub>2</sub>O decomposition mechanism.

### THEORETICAL APPROACH

In this work, a set of heterogeneous reaction mechanisms using the Langmuir & Hinshelwood model [Smiths, 1981] was suggested and it is as follows:



The mechanism (1) is adsorption of N<sub>2</sub>O on CaO surface. The s is an active site on the surface of CaO where N<sub>2</sub>O adsorption and decomposition can occur, and N<sub>2</sub>Os describes the adsorbed state of N<sub>2</sub>O on the active site of CaO. If Eq. (2) is the rate determining step which means the slowest reaction, equation of reaction can be expressed as follows:

$$r = \frac{k_{s1}K_1S[\text{N}_2\text{O}]}{1+K_1[\text{N}_2\text{O}]} = \frac{k'_{s1}K_1[\text{N}_2\text{O}]}{1+K_1[\text{N}_2\text{O}]} \quad (3)$$

Where  $k'_{s1} = k_{s1}S$  and S is the number of total active sites on CaO.

In general, CFBC utilizes the technique of sub-stoichiometric oxygen supply ratio in the lower part of CFBC to control NO<sub>x</sub> emission, CO concentration in this area is usually high [Basu and Fraser, 1991]. CO also can be adsorbed on CaO and it affects the N<sub>2</sub>O decomposition. A bimolecular reaction mechanism is suggested in this work. The heterogeneous mechanism involving the reaction between

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CO and N<sub>2</sub>O on CaO surface is as follows:



When CO coexists with N<sub>2</sub>O then Eq. (5) as well as (2) can be considered for the N<sub>2</sub>O decomposition mechanism. The decomposition reaction rate can be increased with CO. If the mechanism (5) is considered to be the rate determining step and we do not differentiate the N<sub>2</sub>O adsorption site and CO adsorption site, we can suggest following rate equation:

$$r = \frac{k_{s2}SK_1[\text{N}_2\text{O}]K_2[\text{CO}]}{(1+K_1[\text{N}_2\text{O}] + K_2[\text{CO}])^2} = \frac{k'_{s2}K_1[\text{N}_2\text{O}]_o^2(1-x)(\alpha-x)}{(1+K_1[\text{N}_2\text{O}]_o(1-x) + K_2[\text{N}_2\text{O}]_o(\alpha-x))^2} \quad (6)$$

Where,  $x$  is the conversion of [N<sub>2</sub>O],  $\alpha = [\text{CO}]_o/[\text{N}_2\text{O}]_o$ , and  $k'_{s2} = k_{s2}S$ .

After the arrangement we get:

$$A^2 \ln \frac{\alpha-x}{\alpha(1-x)} - 2AB[\text{N}_2\text{O}]_o \ln \frac{(\alpha-x)^\alpha}{\alpha^\alpha(1-x)} = -k'_{s2} \tau (\alpha-1)[\text{N}_2\text{O}]_o \quad (7)$$

Where, [N<sub>2</sub>O]<sub>o</sub> is the concentration of introducing N<sub>2</sub>O,  $A=1+(K_1+K_2\alpha)[\text{N}_2\text{O}]_o$ ,  $B=K_1+K_2$ , and the residence time( $t$ ) is  $\tau=W/\rho F$ . Where  $W$  is mass of CaO bed,  $\rho$  is the particle density,  $F$  is flow rate of gas.

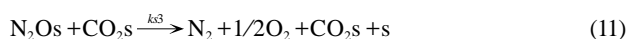
The homogeneous mechanism in this case will be as follows:



The reaction rate will be described as follows:

$$r = k_3[\text{N}_2\text{O}][\text{CO}] \quad (9)$$

CO<sub>2</sub> is considered to hinder the N<sub>2</sub>O decomposition. CO<sub>2</sub> adsorbs on the CaO surface and blocks N<sub>2</sub>O adsorption. CO<sub>2</sub> and N<sub>2</sub>O compete with active site on CaO. But CO<sub>2</sub> does not seem to participate in the N<sub>2</sub>O decomposition reaction. With (1), (10), and (11), the mechanism becomes as follows:



$$r = \frac{k_{s3}S[\text{N}_2\text{O}]}{1+K_1[\text{N}_2\text{O}] + K_3[\text{K}_2\text{CO}_2]} \quad (12)$$

## EXPERIMENTAL

The experimental apparatus used in this study, which is shown in Fig. 1, consists of a reactor, sets of flow meters, a pre-heater, and a set of gas analyzer. The reactor was made of quartz tube with inside diameter of 2.5 cm and height of 50 cm. The middle part of the inside of the reactor is installed with a sintered quartz plate to support the CaO particles. The gas flow is vertically downward through the reactor toward the sintered plate. In order to heat the reactor, an infrared furnace with a PID controller is used. A flow meter was installed at the upstream of gas input in order to regulate gas flow. The reactant gas was pre-heated while passing the inlet tube with a heating band before introduced to the reactor.

Mixtures of CaO particles (0.4 g) and quartz beads (4 g) were loaded on the sintered quartz plate inside the reactor. Nitrogen gas

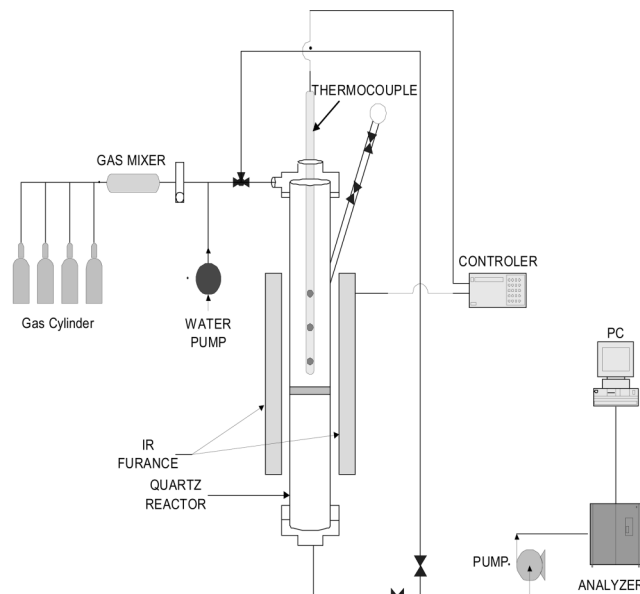


Fig. 1. Diagram of the experimental apparatus.

Table 1. Experimental conditions

Variables	Experimental conditions
Bed materials	CaO (0.4 g)+quartz (4 g)
Flow rate (l/min)	3.5
Temperature (°C)	800-900
N <sub>2</sub> O (ppm)	200-1,000
CO (ppm)	50-3,000
CO <sub>2</sub> (%)	3-20

was introduced into the reactor during the heating of the reactor. When the temperature and other conditions of the reactor reached the preset experimental condition, the reactant gas was introduced into the reactor. The calcium oxide (CaO 54.14%, MgO 0.94%, H<sub>2</sub>O 0.02%) used in the experiment was prepared from the limestone in Chechun province in Korea. It was prepared by calcining at 1,123 K in reactor for 2 hours in N<sub>2</sub> environment. The experimental conditions are listed in Table 1.

## RESULTS AND DISCUSSION

In this study the governing reaction was assumed to be the intrinsic reaction. Therefore, we had to estimate the difference between surface and bulk concentration of N<sub>2</sub>O over CaO particle. When the concentration difference is acceptably small, we can consider that the mass transfer between the bulk and the surface does not affect the total kinetics. The difference between the bulk concentration and the surface concentration can be simply correlated in terms of dimensionless group, i.e., in terms of j-factor [Smith, 1981]. The j-factor for mass transfer is the function of Sherwood number,  $k_m \rho / G$ , the Reynolds number  $d_p G / \mu$ , and Schmidt number  $\mu / \rho D$ . The equation is as follows [Shun et al., 2001];

$$j_D = \frac{k_m \rho a_m}{G} \left( \frac{\mu}{\rho D} \right)^{2/3} \quad (11)$$

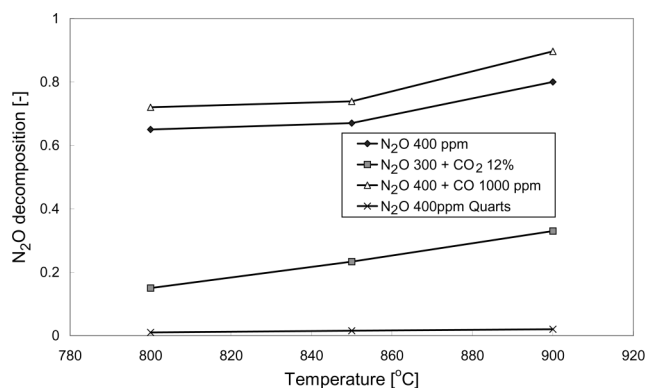


Fig. 2. The effects of temperature on  $\text{N}_2\text{O}$  decomposition with various reactants.

The ratio ( $a_m/a_t$ ) allows for the possibility that the effective mass-transfer area ( $a_m$ ), may be less than the total external area ( $a_t$ ) of the particle. Experimental reaction rate is as follows.

$$r_p = k_m a_m (C_b - C_a) \quad (12)$$

The difference between the bulk concentration and the surface concentration can be calculated by combining Eqs. (11) and (12). In a typical reaction condition  $C_b$  ( $\text{N}_2\text{O}$ ) is  $1.6 \times 10^{-6} \text{ mmol/m}^3$  and calculated  $C_b - C_a$  is  $3.2 \times 10^{-10} \text{ mmol/m}^3$  at  $850^\circ\text{C}$ . Since the concentration  $C_b$  is similar to  $C_a$ , intrinsic reaction rate can be calculated by using the bulk concentration of  $\text{N}_2\text{O}$ . The external diffusion resistance is negligible and the kinetics of  $\text{N}_2\text{O}$  decomposition is mainly the intrinsic kinetics.

Fig. 2 presents the effects of temperature on  $\text{N}_2\text{O}$  (400 ppm) decomposition in  $\text{N}_2$  environment,  $\text{N}_2\text{O}$  (300 ppm) with  $\text{CO}$  (1,000 ppm) decomposition and  $\text{N}_2\text{O}$  (400 ppm) with 12%  $\text{CO}_2$  decomposition, respectively. The thermal effect on  $\text{N}_2\text{O}$  decomposition was also compared by installing quartz sand instead of  $\text{CaO}$ /quartz sand mixture. The thermal effect on  $\text{N}_2\text{O}$  decomposition was negligible. The decomposition of  $\text{N}_2\text{O}$  increased over  $\text{CaO}$  bed with the increase of the temperature. The existence of  $\text{CO}$  further enhanced the  $\text{N}_2\text{O}$  decomposition, whereas  $\text{CO}_2$  hindered the  $\text{N}_2\text{O}$  decomposition over  $\text{CaO}$ . Table 2 listed the surface area before and after the reaction. The surface area of  $\text{CaO}$  did not vary by decomposition of  $\text{N}_2\text{O}$  in  $\text{N}_2$  environment only, but the area was significantly reduced in  $\text{N}_2$  and  $\text{CO}_2$  environment. Thus  $\text{CO}_2$  hinders the  $\text{N}_2\text{O}$  decomposition reaction by adsorption of  $\text{CO}_2$  on  $\text{CaO}$  surface.

Fig. 3 presents the effect of feed concentration of  $\text{N}_2\text{O}$  on its decomposition. The decomposition decreased with the increase of the  $\text{N}_2\text{O}$  concentration due to the relative decrease of the active site on  $\text{CaO}$  surface. A heterogeneous reaction mechanism was proposed and the mechanism as explained in Eqs. (1), (2), (3) showed best fit. The detailed result was reported in the previous paper [Shun et

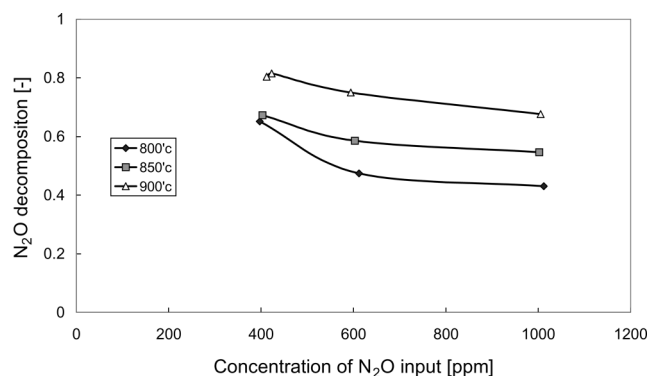


Fig. 3. The effect of concentration on  $\text{N}_2\text{O}$  decomposition.

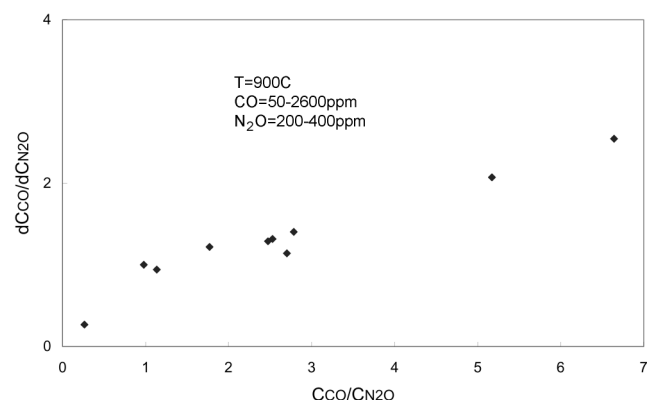


Fig. 4. The effect of feed  $\text{CO}/\text{N}_2\text{O}$  ratio on the decomposition ratio of  $\text{CO}/\text{N}_2\text{O}$ .

al., 2001].

Fig. 4 presents the effect of initial  $\text{CO}/\text{N}_2\text{O}$  ratio on the ratio of  $\text{CO}/\text{N}_2\text{O}$  decomposition. The decomposed mole ratio of  $\text{CO}/\text{N}_2\text{O}$  was almost equal during those of feed  $\text{CO}/\text{N}_2\text{O}$  varying from 1 to 3. That implies when  $\text{CO}$  exists the mechanism of the decomposition of  $\text{N}_2\text{O}$  prefers bi-molecular reaction, and the reaction mechanism of (4) and (5) was proposed. However, further increase of feed  $\text{CO}/\text{N}_2\text{O}$  ratio increased the ratio of decomposition further, and more  $\text{CO}$  is consumed than  $\text{N}_2\text{O}$  is decomposed. This violates the 1 to 1 ratio of the reaction between  $\text{N}_2\text{O}$  and  $\text{CO}$ . It may due to the adsorption of  $\text{CO}$  to  $\text{CaO}$  surface [Shun et al., 2001]. The Fig. 5 and the Fig. 6 compares the heterogeneous mechanism of  $\text{CO}$  and  $\text{N}_2\text{O}$  decomposition as was shown in heterogeneous Eq. (6) and (7) and a homogeneous mechanism (8). Even though the correlation coefficient is low, the heterogeneous model shows the better fit. It seems the simultaneous reaction mechanisms (2) and (5) are competing with each other, and also  $\text{CO}$  is adsorbed on  $\text{CaO}$  surface for  $\text{N}_2\text{O}$  decomposition with the existence of  $\text{CO}$ .

Table 2.  $\text{CaCO}_3$  and  $\text{CaO}$  analyses

Limestone		$\text{CaO}$ , 54.14%; $\text{MgO}$ , 0.94%; $\text{H}_2\text{O}$ , 0.02%
BET surface area [ $\text{m}^2/\text{g}$ ]	Limestone	4.36-4.52
	$\text{CaO}$	7.52-8.02
	After $\text{N}_2\text{O}$ decomposition ( $\text{N}_2$ only)	7.32-7.94
	After $\text{N}_2\text{O}$ decomposition ( $\text{N}_2$ , 80%, $\text{CO}_2$ 20%)	2.35

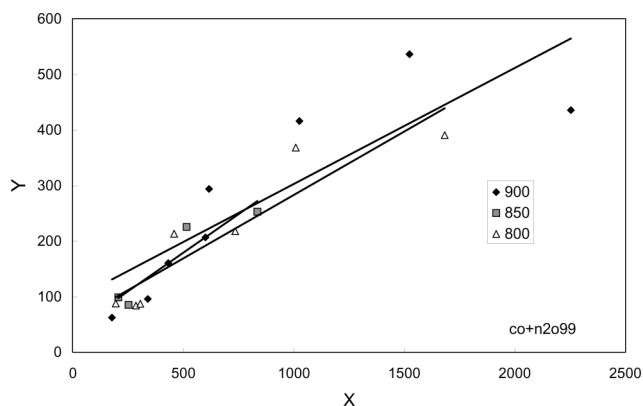


Fig. 5. The heterogeneous mechanism CO and N<sub>2</sub>O decomposition.

$$X = AB[N_2O]_0 \ln \frac{(\alpha-x)^\alpha}{\alpha^\alpha(1-x)} \quad Y = A^2 \ln \frac{(\alpha-x)}{\alpha(1-x)}$$

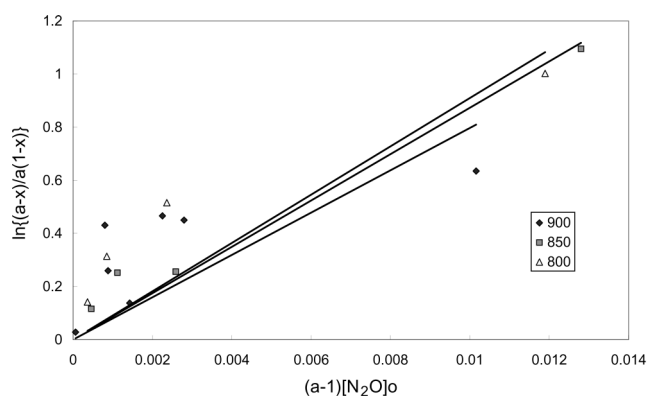


Fig. 6. The homogeneous mechanism of CO and N<sub>2</sub>O decomposition.

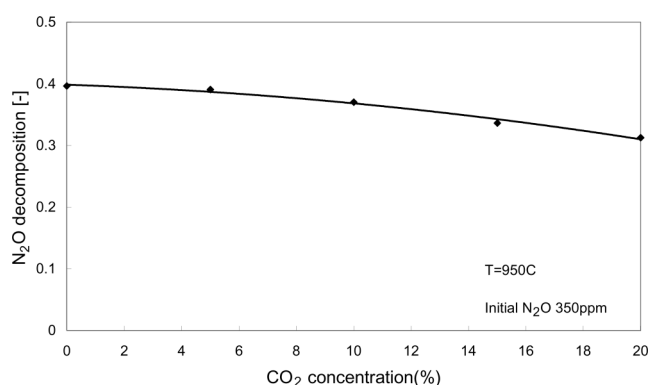


Fig. 7. The effect of CO<sub>2</sub> concentration on N<sub>2</sub>O decomposition.

The influence of CO<sub>2</sub> on N<sub>2</sub>O decomposition over CaO is presented in Fig. 7. As the concentration of CO<sub>2</sub> increased, the N<sub>2</sub>O conversion was gradually decreased until the concentration of CO<sub>2</sub> reached 20%. The variation of N<sub>2</sub>O decomposition was about 10% from 0% to 20% of CO<sub>2</sub>. CO<sub>2</sub> and N<sub>2</sub>O seemed to compete with each other over the active site on CaO, and which resulted in the hindrance of catalyst decomposition of N<sub>2</sub>O on the CaO surface [Chang et al., 2000].

Fig. 8 presents the effect of the initial concentration of N<sub>2</sub>O and

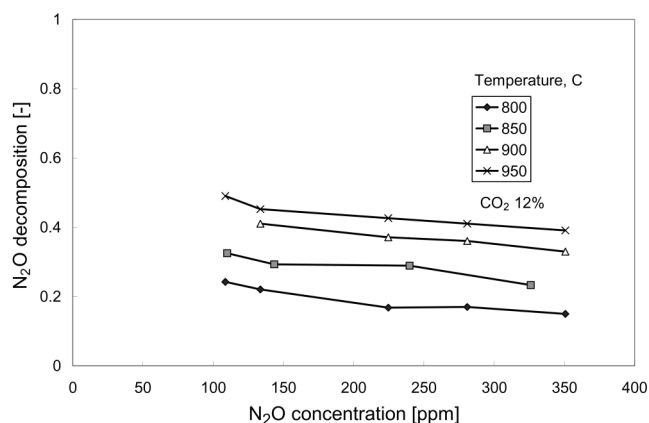


Fig. 8. The effect of feed N<sub>2</sub>O concentration and temperature on its decomposition with CO<sub>2</sub>.

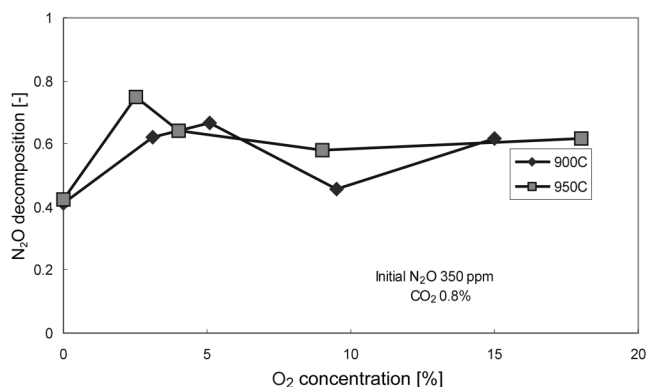


Fig. 9. The effect of O<sub>2</sub> on N<sub>2</sub>O decomposition.

reaction temperature on N<sub>2</sub>O decomposition when the concentration of CO<sub>2</sub> was fixed at 12%. The conversion decreased gradually with the increase of the concentration due to the relative decrease of the CaO surface. However, the conversion increased with the increase of the temperature. That implies the reaction of CO<sub>2</sub> adsorption on the CaO is occurring in the reaction condition, but it decomposes with the increase of the temperature. There is no evidence of direct involvement of CO<sub>2</sub> on N<sub>2</sub>O decomposition, but it still hinders the decomposition. It is concluded that CO<sub>2</sub> competes with the N<sub>2</sub>O on active sites on CaO. Thus, the reaction mechanisms of (10), (11), and (12) were proposed.

The Fig. 9 shows the effect of O<sub>2</sub>, and O<sub>2</sub> also affected N<sub>2</sub>O decomposition. The decomposition increased until O<sub>2</sub> was 2.5–4% and also with the increase of the temperature [Satsuma et al., 2000]. Above the concentration the decomposition rate stabled. With limited information, it seems that O<sub>2</sub> refreshes the CaO surface, but further increase of O<sub>2</sub> partial pressure does not affect either N<sub>2</sub>O adsorption or clearing the CaO surface.

NO also showed the hindering effect in N<sub>2</sub>O decomposition. The effect was, however, relatively low. Fig. 10 shows the effect of NO concentration on N<sub>2</sub>O decomposition. The reaction was monitored with the CO of 10% and 20% concentrations.

In general operation of CFBC, CaO is introduced in the lower part of the combustor. The addition of CaO in the lower part of the combustor will decrease N<sub>2</sub>O formation; however, it increases NO

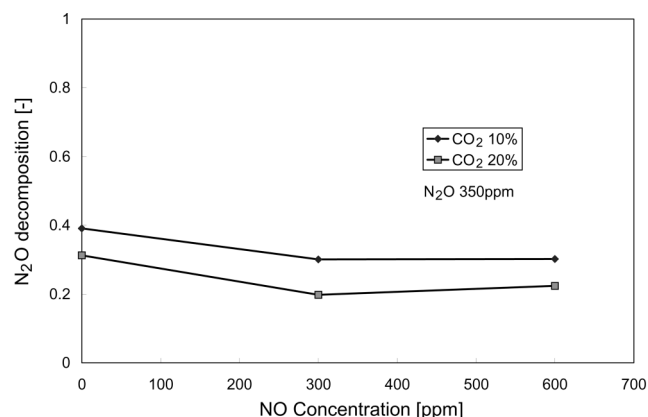


Fig. 10. The effect of NO concentration on N<sub>2</sub>O decomposition.

emission [Shimizu, 2000]. In order to accomplish the simultaneous reduction of both NO and N<sub>2</sub>O in commercial CFB boilers, the supply of limestone which can reduce N<sub>2</sub>O at the lower part of the combustor will be a measure for N<sub>2</sub>O control; that and typical SNCR (selective non catalytic reaction) techniques can be applied at the flue gas stage [Ljungdahl, 2001] to remove NO with increased with the injection of limestone.

### CONCLUSION

The conclusions of N<sub>2</sub>O decomposition on the surface of calcium oxide are as follows.

1. CaO from domestic limestone showed catalytic activity on N<sub>2</sub>O decomposition.
2. A set of heterogeneous mechanisms of N<sub>2</sub>O decomposition with CO and CO<sub>2</sub> were proposed from the experimental data.
3. CO promoted N<sub>2</sub>O decomposition.
4. CO<sub>2</sub> hindered N<sub>2</sub>O decomposition and the increased CO<sub>2</sub> concentration gradually decreased N<sub>2</sub>O decomposition.
5. O<sub>2</sub> increased N<sub>2</sub>O decomposition up to 3%; then above the concentration the effect was not clear.
6. NO also hindered N<sub>2</sub>O decomposition.
7. In order to reduce N<sub>2</sub>O and NO simultaneously in the CFBC combustor, feeding of CaO in the lower part of the combustor is recommended. The increased NO should be removed from flue gas by SNCR method.

### NOMENCLATURES

- $r$  : reaction rate  
 $K$  : equilibrium constant  
 $k_s'$  : reaction rate constant [1/s]  
 $S$  : total active sites existing in the surface of CaO  
 $F$  : flow rate [m<sup>3</sup>/s]  
 $X_i$  : conversion of  $i$  component  
 $T$  : temperature [K]  
 $W$  : packed weight of CaO [kg]  
 $[i]_0$  : initial concentration of  $i$  component  
 $\alpha$  : ratio of CO and N<sub>2</sub>O concentration  
 $\rho$  : density of CaO particle [kg/m<sup>3</sup>]

- $\tau$  : residence time [s]  
 $\mu$  : gas viscosity [kg/m-s]  
 $D$  : molecular diffusivity [m<sup>2</sup>/s]

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