A Study on the N$_2$O Reduction Rate According to Temperature and Residence Time in the Exhaust Gas Atmosphere Emitted on Combustion of Air and Oxygen

Jae Geun Yun, Ji Yeop Kim, Han Min Lee, Gwang Yeol Baik, Min Kyu Jeon, Jin Han Yun, and Jung Goo Hong*

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ABSTRACT: N$_2$O is a hazardous greenhouse gas. It should be reduced to solve global warming problems. In this study, experiments of N$_2$O thermal decomposition were conducted by simulating the exhaust gas atmosphere emitted during the combustion of air and pure oxygen in an actual circulating fluidized bed system and incinerator system. As a result of comparing the N$_2$O reduction rate in N$_2$ and CO$_2$ atmospheres, the N$_2$O reduction rate in the CO$_2$ atmosphere was 20% higher than that in the N$_2$ atmosphere. It is judged that the N$_2$O reduction rate is high in a CO$_2$ atmosphere (exhaust gas from pure oxygen combustion) due to complex factors such as the reverse reaction, the diffusion coefficient, and static pressure-specific heat. Therefore, pure oxygen combustion increases the reduction rate of nitrous oxide. In addition, when operated with an appropriate residence time and temperature, a reduction effect of more than 95% can be expected, and the fuel consumption rate is also expected to improve.

1. INTRODUCTION

Climate change due to global warming is emerging as a major environmental problem. A critical cause of global warming is greenhouse gases. Greenhouse gases include CO$_2$, CH$_4$, N$_2$O, hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), and SF$_6$. In particular, N$_2$O is exhaust gas from waste incinerators. It is emitted by anthropogenic factors and causes ozone layer destruction in the stratosphere, but unlike nitrogen oxide (NOx), it has been excluded from strict regulation. In addition, N$_2$O has a global warming index that is 310 times higher than that of CO$_2$ and is a substance that destroys the earth’s ozone layer. Further, N$_2$O is known to be mostly generated from fixed-emission sources such as industrial boilers, large power plants, and waste incinerators.$^{1−4}$ Oxy-combustion technology is employed for capturing, using, and storing CO$_2$. It uses O$_2$ separated by an air separation unit and CO$_2$ from the combustion exhaust gas. In the process of oxy-fuel circulating fluidized bed combustion (Oxy-CFBC), N$_2$O may be generated depending on the conditions in the furnace, and it does not cause visible air pollution problems such as ultrafine dust. Therefore, little is known about the severity of the N$_2$O gas emissions. The main emission source for N$_2$O may be generated by the coal surface combustion reaction in coal-fired power plants and the side reaction of the selective catalytic reduction (SCR) process of NOx.$^5$ A study was conducted on the behavior of pollutants according to the application of desulfurization and denitrification methods in a pure-oxygen circulating fluidized bed.$^6$ The concentration of NO in the gas decreased from 130 to 34 ppm, whereas the concentration of N$_2$O tended to increase from 61 to 156 ppm. In addition, it was reported that N$_2$O formation occurred during the oxidation of N$_2$ contained in fuel under pure oxygen combustion conditions.$^6$

SCR and selective noncatalytic reduction (SNCR) are commonly used as processes for controlling NOx emitted from waste incinerators. SCR uses a catalyst and controls the exhaust gas. The processing efficiency of SCR is high, and it can be processed at a relatively low temperature of 500−600 K. In addition, although it has the advantages of easy maintenance and repair, there are disadvantages such as the restriction of the exhaust gas temperature, expensive installation costs, expensive catalysts, fuel costs, and corrosion phenomena. SNCR has a lower treatment efficiency than SCR, but the initial installation cost is low, and it can be easily installed in a short period.

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Many incinerators have applied SNCR. In addition, SNCR reduces NOx to N2 and water vapor by spraying ammonia or urea at a furnace temperature of 1173−1373 K to reduce NOx generated during combustion. However, it is problematic that the reduced NOx is not converted to N2 but re-emitted as N2O.10,11

In addition, in a study on the N2O emission characteristics and emission coefficients of solid waste incineration facilities, Choi and Ko9 measured the N2O concentration for 24 h three times: 114.8, 125.4, and 134.7 ppm, respectively; the average operating temperature was 1273 K, and the average oxygen concentration was measured to be 5−7%.

Studies have reported that the lower the oxygen concentration among the operating conditions of the waste incineration facility, the higher the NOx generation and the lower the N2O generation rate.10,11 In addition, it was reported that the decomposition mechanism is temperature-dependent because N2O is highly sensitive to temperature.12 In a study on the thermal decomposition and reaction rate of N2O in a high-temperature inert gas atmosphere, Lee et al.13 reported that most reduced N2O was converted to N2.

In a study on the generation and decomposition of N2O/NO according to the reactor temperature and residence time in an Ar atmosphere, Lee et al.14 reported that an appropriate residence time was required for decomposing N2O.

Liu et al.15 reported that enormous O radicals are generated during CO combustion, which promotes N2O decomposition. Kilpinen and Hupa16 reported HCN as an excellent material for forming N2O; HCN converts to intermediate NCO under lean fuel atmosphere conditions, and N2O is produced by the reaction of NCO with NO.

Clearly, N2O must be reduced as a greenhouse gas. Through various previous studies, it was confirmed that N2O is sensitive to temperature, and most of it is thermally decomposed by temperature. The residence time is also an essential operating parameter in SNCR. In addition, it was confirmed that most N2O was decomposed at 1273 K in an Ar atmosphere, and the longer the residence time, the better the reduction effect. The residence time is the residence time of the exhaust gas from the incinerator to the inlet of the waste heat boiler. A residence time that is too long cannot be applied to actual operating conditions. Further, it is crucial to find an appropriate residence time. Therefore, in this study, the exhaust gas atmosphere and SNCR during actual air combustion and pure oxygen combustion are simulated. In addition, we conduct an N2O decomposition experiment according to temperature and residence time in an environment that simulates the process in which the actual exhaust gas is emitted. Through experimentation, we seek the appropriate temperature and residence time in the exhaust gas environment emitted from the air and pure oxygen and in SNCR. Further, we verify the N2O decomposition rate in each atmosphere. In addition, N2O decomposition reaction equations are obtained using a reaction path analyzer (RPA), and the decomposition characteristics of N2O in each atmosphere are identified. If there is a difference in the N2O reduction rate depending on the atmosphere, the cause is probed. In particular, we examine the difference in the reduction rate depending on the atmosphere through the intrinsic properties of the molecules, such as the reverse reaction, the diffusion coefficient, and static pressure-specific heat.

2. RESULTS AND DISCUSSION

2.1. Thermal Decomposition of N2O According to Residence Time and Temperature in an N2 Atmosphere

Figure 1 shows the N2O reduction rate with respect to the change in the reactor temperature and residence time in an N2 atmosphere (exhaust gas atmosphere discharged during air combustion). The N2O reduction rate was expressed as a ratio of the input N2O concentration of 400 ppm to the reduced N2O concentration after the reaction, as shown in eq 1.

\[
\text{N}_2\text{O reduction} (%) = \frac{\text{reduced } \text{N}_2\text{O concentration}}{\text{inlet } \text{N}_2\text{O concentration}} \times 100
\]

(1)

Nine experiments were conducted by changing the residence time to 7, 10, and 13 s and simultaneously increasing the temperature to 1073, 1173, and 1273 K in an N2 atmosphere. First, we discuss the temperature. At a temperature of 1073 K based on a residence time of 7 s, the N2O reduction rate was only 10%. However, in the temperature range of 1173 and 1273 K, the reduction rates were 39 and 91%, respectively. A similar trend was also observed at residence times of 10 and 13 s. Accordingly, it was confirmed that most N2O decomposed under high temperatures, even in an N2 atmosphere. Next, we consider the residence time. The difference in the N2O reduction rate with respect to the residence time based on 1073 and 1273 K was insignificant, whereas it was significant for 1173 K. In other words, the N2O decomposition at an average temperature of 1173 K is significantly affected by the residence time. Through the experiment results, we can confirm how much the residence time affects the N2O decomposition in SNCR. Therefore, it was confirmed that N2O was significantly affected by temperature and residence time even in an N2 atmosphere. Figure 2 was calculated through CHEMKIN-PRO with a temperature of 1073 K and a residence time of 7 s. Figure 2 shows the reactants remaining in one temperature zone at the end of the reactor. It shows that it was possible to check the gas components at the rear end of the reactor after the reaction. In addition, the remaining components after N2O input being O radicals as intermediates and N2 and NOx as final byproducts were confirmed.

Figures 3−5 show the specific decomposition mechanism of N2O and the reaction rate of each reaction formula obtained.
from the CHEMKIN-PRO RPA. It shows that most N2Oi s
reduced to N2 by eqs 2 and 3. In this study, a reduction
pathway decomposed from N2O to NO was also discovered,
but simulation results were not attached because this reaction
accounted for less than 2% of the overall reaction. Tables 1 and
2 show the specific reaction rate (rate of progress) values
plotted in Figures 3−5.

In particular, it was confirmed that the reaction continued to
occur according to eq 2 as the temperature and residence time
increased. In addition, the numerical value of the reaction rate
decreased as the temperature and residence time increased.
The unit of the reaction rates is mol/s, and the chemical
reaction rate is affected by the concentration of the reactants.
As the concentration of N2O increased, the number of
intermolecular collisions increased. The concentration
decreased due to intermolecular collisions. As the concentration
decreased, the corresponding reaction rate value also
decreased. Therefore, as the concentration decreased, the
number of molecules per unit volume decreased, thereby
reducing collisions between molecules; accordingly, the
chemical reaction and reaction rate decreased. Conversely,
when the concentration of N2O measured at the rear end of
the reactor was high, the reaction rate values of eqs 2 and 3
would be high. Moreover, it was confirmed through
calculations that the reaction rate decreased with the increase
in residence time under the same temperature. As the
residence time of the gas increased, the probability of
molecular collision increased. Therefore, it was judged that
the concentration of N2O discharged from the rear end of the
reactor was reduced because it had a sufficient chemical
reaction between molecules, which was similar to the trend of
the concentration value of N2O measured at the rear end of the
actual reactor. Therefore, N2O was mostly decomposed by
heat regardless of the composition of the exhaust gas.

\[ \text{N}_2\text{O}(+\text{M}) \leftrightarrow \text{N}_2 + \text{O}(+\text{M}) \]  \hspace{1cm} (2)
\[ \text{N}_2\text{O} + \text{O} \leftrightarrow \text{N}_2 + \text{O}_2 \]  \hspace{1cm} (3)

2.2. Thermal Decomposition of N2O According to
Residence Time and Temperature in a CO2 Atmos-
phere. Figure 6 shows the N2O reduction rate with respect to
the change in the reactor temperature and residence time in a
CO2 atmosphere (exhaust gas atmosphere discharged during
oxygen combustion). Except for the gas atmosphere, other
experimental conditions are the same as the previous
experimental conditions. First, we consider the temperature.
At a temperature of 1073 K based on a residence time of 7 s,
the N2O reduction rate was 35%. In the temperature range of
1173 and 1273 K, the reduction rates were 60 and 90.4%,
respectively. It was confirmed that the respective reduction
rates of 1073, 1173, and 1273 K were 36, 73, and 94% based
on the residence time of 10 s, indicating similar trends. Based
on the residence time of 13 s, the reduction rates for
temperatures of 1073, 1173, and 1273 K were 38, 81, and 94%,
respectively, which are similar to the previous experimental
results. In the CO2 atmosphere, the N2O reduction rate
increased with the temperature.

From Figure 6, N2O was mostly decomposed at high
temperatures in the CO2 atmosphere. Next, we consider the
residence time. The difference in N2O reduction rate with
respect to the residence time at 1073 and 1273 K for the residence time of 7 s did not change significantly, whereas it increased significantly at 1173 K. At this time, it can be confirmed how much the residence time affects the decomposition of N2O in SNCR. Therefore, it was confirmed that N2O was significantly affected by the operating temperature and residence time of SNCR. In particular, when the residence time was increased in a CO2 atmosphere, N2O could be reduced by more than 90%, even at the early 1200 K in SNCR. It was judged that this would significantly influence fuel use efficiency, management, and maintenance in SNCR. However, a long residence time of 10 s or more was considered difficult to apply to the actual process. Therefore, an appropriate operating temperature and residence time should be employed.

Figure 7 was calculated through CHEMKIN-PRO with a temperature of 1073 K and a residence time of 7 s. It shows that it was possible to check the gas components at the rear end of the reactor after the reaction. In addition, NCO, an intermediate, was generated from the remaining components after N2O input, which differed from the results in the N2 atmosphere, and N2, NOx, and CO were generated as final reactants.

Figure 7. Schematic of N2O decomposition mechanism at a residence time of 7 s in a CO2 atmosphere.

Figures 8–10 show the specific decomposition mechanism of N2O and the reaction rate of each reaction formula obtained from the CHEMKIN-PRO RPA. In the CO2 atmosphere, most N2O is reduced to N2 by eqs 2 and 3. Tables 3 and 4 show the specific reaction rate (rate of progress) values plotted in Figures 8–10.

The numerical value of the reaction rate decreased as the temperature and residence time increased by Figures 8–10. In addition, it was confirmed through calculations that the value of the reaction rate decreased with an increase in residence time under the same temperature. The trend was similar to the...
Therefore, in this study, it was judged that the reverse reaction of eqs 4 and 5 contributed to some extent in increasing the reduction rate in the CO₂ atmosphere.

2.3. Comparison of N₂O Reduction Rates in N₂ and CO₂ Atmospheres. Figure 12 depicts a graph comparing the N₂O reduction rates of the N₂ and CO₂ atmospheres at a residence time of 7 s. The reduction rate of the CO₂ atmosphere at 1073 K was 24% higher than that of the N₂ atmosphere. In addition, at 1173 K, the reduction rate in the CO₂ atmosphere was 20% higher than that of the N₂ atmosphere. Table 1 shows the diffusion coefficient values of N₂ and CO₂. Figure 13 shows a diagram explaining the correlation between the diffusion coefficient and the number of molecules.

Table 5 shows that CO₂ has two times the diffusion coefficient of N₂. Figure 13 shows that there were more CO₂ molecules than N₂ in the same area. Therefore, it was judged that the number of molecular collisions between N₂O and CO₂ injected into the reactor increased, contributing to an increase in the N₂O reduction rate.

Table 6 shows the constant pressure-specific heat table of the gas used in the experiment. In this study, CO₂ and N₂ were used because their atmospheric pressures were constant. The atmospheric pressure of CO₂ was lower than that of N₂. In this study, it was judged that the constant heat-specific pressure contributed the most to the increase in the N₂O reduction rate in the CO₂ atmosphere. Specific heat is defined as the amount of heat required to raise the temperature of 1 kg of a substance by approximately 273 K. The diffusion coefficient differed depending on the temperature.

Therefore, in this study, it could not be judged that the reduction rate increased only by the diffusion coefficient. Because the temperature of CO₂ was higher than that of N₂ when the heater of the reactor was at the same temperature, through the experimental results, it was confirmed that most N₂O decomposed at high temperatures. When the temperature of the atmosphere increased, the molecular motion of N₂O was active, and the number of collisions increased. As the number of collisions increased, the concentration of molecules decreased. Therefore, it was judged that CO₂, which had a high heated temperature compared with N₂, had the most significant influence on the N₂O reduction rate.

3. CONCLUSIONS

In this study, experiments of N₂O thermal decomposition were conducted by simulating the exhaust gas atmosphere emitted during the combustion of air and pure oxygen in an actual waste incinerator system.

Table 3. Rate Progress of eq 2 between N₂O and N₂ in a CO₂ Atmosphere

| Temperature (K) | 7 s | 10 s | 13 s |
|-----------------|-----|------|------|
| 1073 K          | forward reaction rate: 1.06 × 10⁻¹⁰; reverse reaction rate: −1.28 × 10⁻¹⁶ | forward reaction rate: 8.95 × 10⁻¹¹; reverse reaction rate: −1.38 × 10⁻¹⁶ | forward reaction rate: 7.65 × 10⁻¹¹; reverse reaction rate: −1.39 × 10⁻¹⁶ |
| 1173 K          | forward reaction rate: 7.25 × 10⁻¹¹; reverse reaction rate: −4.03 × 10⁻¹⁶ | forward reaction rate: 2.65 × 10⁻¹¹; reverse reaction rate: −1.7 × 10⁻¹⁶ | forward reaction rate: 9.91 × 10⁻¹²; reverse reaction rate: −7.54 × 10⁻¹⁶ |
| 1273 K          | forward reaction rate: 2.55 × 10⁻¹⁵; reverse reaction rate: −1.03 × 10⁻¹⁶ | forward reaction rate: 1.95 × 10⁻¹⁶; reverse reaction rate: −1.03 × 10⁻¹⁶ | forward reaction rate: 9.81 × 10⁻¹⁷; reverse reaction rate: −1.03 × 10⁻¹⁶ |
The concentration of N₂O measured at the rear end of the reactor decreased.

(2) Through the experimental results, it was confirmed that N₂O was affected by temperature and residence time regardless of the atmosphere of most exhaust gases generated during actual combustion. Accordingly, appropriate operating conditions (residence time and temperature) are required. If the residence time is increased, combustion is possible in the low-temperature region. Therefore, an increase in the residence time may increase the fuel efficiency.

(3) Compared to the exhaust gas of air combustion, the reduction rate of nitrous oxide in the exhaust gas environment of pure oxygen combustion was about 20% higher. Through a series of experimental results, it is judged that pure oxygen combustion rather than air combustion is an effective method for reducing N₂O. In addition, if the appropriate residence time and temperature are adjusted, the reduction rate of N₂O is judged to be close to 95% or more.

4. MATERIALS AND METHODS

4.1. Experimental Setup. Figures 14 and 15 show a schematic and a picture, respectively, of a laboratory-scale electric furnace reactor for N₂O thermal decomposition. The experimental apparatus comprises a gas supply unit, a thermal reactor, a measurement device, and an emission part. Experimental gases (N₂, CO₂, O₂, and N₂O) flow through a mass flow controller. The thermal reactor is a cylindrical tube (inner diameter: 114 mm; length: 750 mm) and can be heated up to 1373 K using three internal heating coils. An insulator surrounded the reactor to prevent heat loss and maintain the temperature of the experiment. The temperature of the reactor was measured using R-type thermocouples. The concentration of the exhaust gas after the reaction was measured using a nondispersive infrared gas meter (NOVA IR-Pro). The measurement error of the gas meter was below 2% (electrochemical sensor).

4.2. Experimental Conditions. Table 7 shows the composition ratio of the gas mixer used in the N₂O pyrolysis
experiment. The gas mixer comprised N\textsubscript{2} (94.96%), CO\textsubscript{2} (94.96%), O\textsubscript{2} (5%), and N\textsubscript{2}O (400 ppm) as conditions for simulating the exhaust gas from SNCR. The actual combustion exhaust gas composition was the N\textsubscript{2} and CO\textsubscript{2} atmospheres. The residence time is the time the mixed gas stays in the furnace. In this study, the residence time was set to about 7 s in consideration of the time the mixed gas moved from the combustion chamber in the incinerator to the inlet of the waste heat boiler. The mixer flow rate corresponding to each residence time was analyzed by equation 6 using the reactor volume and residence time. Here, \( Q \) is the mixer flow rate, \( D \) is the inner diameter of the reactor, \( L \) is the length of the reactor, and \( t \) is the residence time.

\[
Q = \frac{\pi \times (D/2)^2 \times L}{t}
\]  

(6)

Table 8 lists the experimental conditions. Considering the normal SNCR temperature, the experimental temperature ranged from 1073 to 1273 K at a 100 K interval. For a fixed residence time of the gas mixture at each temperature, the flow rate was varied according to Charles’ law, which is expressed as follows:

\[
\frac{Q_1}{T_1} = \frac{Q_2}{T_2}
\]  

(7)

Here, \( Q_1 \) is the inlet flow rate, \( T_1 \) is the inlet temperature, \( Q_2 \) is the flow rate for each temperature, and \( T_2 \) is the reactor temperature. The concentration of the exhaust gas was measured in a 1 min interval at the rear end of the reactor after a 15 min gas supply. The numerical and graphical data reported here are the average values of 10 measurements for each experimental condition. Ambient conditions were maintained in atmospheric air (298 K, 1 atm).

4.3. CHEMKIN Calculation Conditions. The N\textsubscript{2}O thermal decomposition was simulated using CHEMKIN\textsuperscript{17}. A 1D plug flow reactor (PFR) model was employed in the simulations. The PFR model is a cylindrical reactor model that assumes a steady state during flow. The inlet gas was assumed to be perfectly mixed; therefore, continuous thermal reactions were observed inside the reactor. The furnace model had an inner diameter of 114 mm and a length of 750 mm, which was the same as the experimental setup. The temperature of the reactor was divided into three sections ranging from 1073 to 1273 K.
1273 K. The residence time of the gas mixture was set to 7 s considering the actual experimental conditions. The mechanism calculated in this study is the GRI-Mech 3.0.18

GRI-Mech 3.0 is an optimized mechanism designed to model natural gas combustion, including NO formation and reburn chemistry. It is designed to provide solid fundamental kinetics that provides the best-coupled modeling predictability for basic combustion properties. It has 325 reaction mechanisms. The reason for using GRI-Mech 3.0 in this study is that it is a proven mechanism in all cases. Also, GRI-Mech 3.0 was created based on NASA’s thermal data. It was also mostly consistent with the experimental results. The waste incineration system simulated in this study uses methane combustion as a fossil fuel. Therefore, the GRI-Mech 3.0 is judged to be almost as correct as an N₂O-related mechanism. In addition, similar trends were already confirmed in the experimental and CHEMKIN simulation results. Therefore, in this study, the simulation results were judged to be meaningful in the paper and were not attached separately.

Reaction path analysis is not uniform throughout the reactor. This is a natural result. As mentioned in the text, the attached reaction path analysis is the result of checking at the end of the reactor. Naturally, the reaction pathway analysis changes over time as the chemical reactants and products pass through a section with a total length of 750 m. In this study, only the parts to be checked were compared and checked. Thus, it identifies the case where a reactant or product is decomposed and produced in a specific region.

■ AUTHOR INFORMATION

Corresponding Author
Jung Goo Hong — School of Mechanical Engineering, Kyungpook National University, Bukgu, Daegu 41566, Republic of Korea; [email: ighong70@knu.ac.kr; Fax: 82-53-930-6550

Authors
Jae Geun Yun — School of Mechanical Engineering, Kyungpook National University, Bukgu, Daegu 41566, Republic of Korea
Je Yeo Kim — School of Mechanical Engineering, Kyungpook National University, Bukgu, Daegu 41566, Republic of Korea
Han Min Lee — School of Mechanical Engineering, Kyungpook National University, Bukgu, Daegu 41566, Republic of Korea
Gwang Yeol Baik — School of Mechanical Engineering, Kyungpook National University, Bukgu, Daegu 41566, Republic of Korea
Min Kyu Jeon — Department of Environmental Machinery, Korea Institute of Machinery & Materials, Yuseong-gu, Daejeon 34103, Republic of Korea
Jin Han Yun — Department of Environmental Machinery, Korea Institute of Machinery & Materials, Yuseong-gu, Daejeon 34103, Republic of Korea

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.1c05742

Notes
The authors declare no competing financial interest.

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