EFFECT OF ETHENE (C₂H₄) ON THE PLASMA DeNO_x PROCESS FROM DIESEL ENGINE EXHAUST

Kwang-Seo Park^{1)*}, Dong-Inn Kim²⁾, Hyeong-Sang Lee²⁾, Bae-Hyeock Chun³⁾, Woong-Sup Yoon²⁾ and Kwang Min Chun²⁾

¹⁾Dongyang Technical College, 62-160 Gochuk-dong, Guro-gu, Seoul 152-714, Korea ²⁾Yonsei University, 134 Shinchon-dong, Sodaemoon-gu, Seoul 120-749, Korea ³⁾Blue Planet Ltd., YERC 131A, 134 Shinchon-Dong, Seodaemoon-Gu, Seoul 120-749, Korea

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ABSTRACT-Effect of ethene on the $DeNO_x$ conversion process in a simulated diesel engine operating conditions was investigated experimentally and theoretically. With the addition of even a small amount of ethene the NO to NO₂ conversion enhances greatly. The energy required to convert one NO molecule is 27 eV with 250 ppm ethene added, while 137 eV without ethene at 473 K. The effect of energy density, temperature, and the initial concentrations of ethene and oxygen are also discussed and the results show that the increase of the mentioned parameters lead to the promotion of NO oxidation. A kinetic model used in this study shows good agreement with the experimental result. Byproducts like formaldehyde (CH₂O) and methyl nitrite (CH₃ONO) predicted by model calculation are broken up into CO and H₂O eventually when high energy is delivered to the gas mixture. Sensitivity analysis shows that the main reactions of NO oxidation when ethene is added are: $HO_2+NO\rightarrow NO_2+OH$, $RO_2+NO\rightarrow NO_2+RO$, where R is a hydrocarbon radical. Also the direct oxidizing reaction of NO with O cannot be neglected.

KEY WORDS: Plasma, NO_x, Ethene, Diesel engine, Exhaust, NO conversion, Kinetic model, Sensitivity analysis

1. INTRODUCTION

Plasma processing for simultaneous remediation of NO_x and particulates from diesel engine exhaust is currently receiving considerable attention not only because it has a potential of saving aftertreatment expenses but also because this approach has potentially broad impact on the reduction of harmful pollutants from many other sources like lean burn gasoline engines, incinerators, and power plants.

It is by now widely accepted that plasma/catalyst system can remove NO_x efficiently even in oxygen rich exhaust conditions. Though catalytic mechanism of NO_x reduction in a plasma is not clear yet, many experimental and theoretical works show that non-thermal plasma in the presence of oxygen and hydrocarbon will effectively convert NO to NO_2 and the NO_2 can be converted to benign gases of N_2 and O_2 with the correct choice of catalyst. While Hoard and Balmer (1998) demonstrated a proprietary catalyst in combination with the plasma that reduced NO but not NO_2 to N_2 , many researchers have shown that NO_2 can be converted to N_2 more efficiently

than NO on various catalytic materials Penetrante *et al.*, 1998; Chun *et al.*, 2000; Lee *et al.*, 2000; Tonkyn *et al.* (2000) Penetrante *et al.* (1999) also suggested that NO_2 could be used to oxidize the carbon fractions of diesel particulates. Therefore NO_2 plays an important role in the simultaneous removal of NO_x and particulates.

The addition of hydrocarbon has been shown to be an effective way of converting NO to NO₂ with plasma assistance at low temperature and even without plasma assistance at high temperature. Marinov *et al.* (1998) studied the promotion effect of hydrocarbons on the NO to NO₂ conversion at temperatures from 600K to 1100K. The experimental results indicate that, in general, ethene and propane effectively oxidize NO to NO₂ while methane is less effective. But, the effectiveness of hydrocarbons varies appreciably with temperature and no other hydrocarbons except propane show any promotion effect of NO to NO₂ conversion below 700K.

With plasma assistance, many researches have been done with propene since it is known to have a very good NO_x conversion effect. Penetrante *et al.* (1997) observed that about 90% of NO was converted at 20J/L with the addition of 1000 ppm propene, while less than 20% of NO was converted without hydrocarbon even at the high

^{*}Corresponding author. e-mail: kspark@dongyang.ac.kr

energy density. Background gases were 10% O_2 , base N_2 with initial NO concentration of 500 ppm and the temperature was 300°C. Park *et al.* (2000) also proposed that 50% of NO could be converted to NO₂ at 20J/L with 600 ppm propene in a mixture of 10% O_2 , base N_2 with 400 ppm of initial NO concentration and the temperature of 200°C.

Vogtlin and Penetrante (1993) suggested that hydrocarbon additive served by recycling the hydroxyl radicals during the oxidation and reduction of NO and the efficiency of a particular hydrocarbon was therefore limited by the reaction rate of this hydrocarbon with hydroxyl radicals. Their experimental data showed that 18 eV and 29 eV were needed to remove one NO and NO_x molecule, respectively, with the injection ratio of noctane to initial NO_x of 8:1 at 224°C. Sathiamoorthy et al. (1999) has found that 82 eV was needed to remove one NO molecule with the addition of 500 ppm of ethene in dry air with 100 ppm NO at room temperature, a residence time of 14s, and a plasma peak voltage of 49 kV. Mok and Nam (1999) observed that the energy requirement to convert one NO molecule could be reduced to 22 eV and 15 eV with the addition of ethene and propene, respectively, when 60% of the initial NO was converted at room temperature.

The present study examines how ethene converts NO to NO₂ efficiently in a non-thermal plasma. The ethene is chosen to be the hydrocarbon additive since not only it has a good NO_x conversion effect but also it is the major component of diesel exhausted volatile organic compounds (Jaecker-Voirol, 2000). We have performed experiments to quantify the NO to NO₂ conversion and the consumption of ethene in the plasma for gas mixtures containing various levels of oxygen and ethene, base N₂. The temperatures considered are 293K and 473K to simulate the diesel engine operation conditions. The calculation results of chemical kinetics modeling are compared with the experimental data. The model is useful in anticipating byproduct formation that cannot be easily measured in the experiments.

2. EXPERIMENATAL SETUP

Figure 1 shows a schematic diagram of experimental setup used in this study. The pulsed corona discharge reactor consists of a stainless steel wire (ϕ 1.4 mm) inside a stainless steel cylinder (ϕ 36 mm × 280 mm). The inner electrode wire is connected to a pulsed DC high-voltage generator, which has the pulse duration of 1ms with variable voltage and pulse repetition rates up to the kilohertz. The electrical energy deposition into the gas is determined by monitoring the voltage and current waveforms with a digital oscilloscope (LeCroy 140) using Tektronix P6015 high-voltage probe and a A6302/A503B

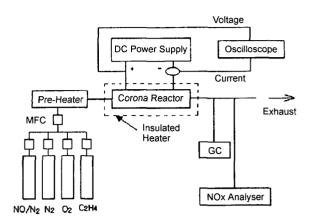


Figure 1. Schematic diagram of experimental setup.

current probe/amplifier. A computer data acquisition system reads the voltage and current profiles and integrates the product of the voltage and discharge current over the pulse duration to yield the pulse energy.

The reactant gas mixtures consist of NO/N₂, N₂, O₂ and C₂H₄. These gases are controlled by MFC (mass flow controller, Brooks 5850E and 5850I), which permits exact control of the flow rate of each mixture component. The gas flow rate is fixed at 1 L/min throughout the test runs. The gas mixture passes through the pre-heater and the reactor banded with heating coils to maintain the target temperatures (293K, 473K). The input energy density, Joules per standard liter (J/L), which is the ratio of discharge power to gas flow rate at standard conditions (25°C and 1 atm) is controlled by changing the pulse repetition rate from 30 Hz to 140 Hz.

The gas composition is analyzed by a chemilumine-scent NO_x analyzer (Thermo Environmental Instruments, Model 42h) for determining NO and NO_x levels, and by a FID gas chromatography (YoungLin M600D) for determining C_2H_4 level. The difference between NO and NO_x readings is taken to be NO_2 .

3. MODELING

Positive pulsed corona discharge in a gaseous medium creates high energetic electrons. Even though the electrons are short-lived under atmospheric pressure conditions and rarely collide with the pollutant molecules, they undergo many collisions with the dominant background gas molecules, thus producing radicals through electronimpact dissociation and ionization. These radicals are chemically active and react with other gas molecules, resulting in the removal of pollutants (Penetrante *et al.*, 1995).

The electron impact dissociation processes considered in this study are as following,

$$e+N_2 - \xrightarrow{k_1} e+N+N$$
 (R1)

$$e+N_2 - \frac{k_2}{} \Rightarrow e+N_2A$$
 (R2)

$$e+O_2 \xrightarrow{k_3} e+O+O$$
 (R3)

$$e+O_2 - \xrightarrow{k_4} e+O+O(^1D)$$
 (R4)

$$e+O_2 \xrightarrow{k_5} e+O_2^*$$
 (R5)

where the k_1 – k_5 are electron collision rate constants. The electron collision rate constant, k, and the electron drift velocity, v_d , both depend on E/N (reduced electric field, E: electric field, N: gas number density) and are calculated by using ELENDIF software (Morgan and Penetrante, 1990), which solves the Boltzmann transport equation. Electron collision cross-sections for each species necessary to run ELENDIF are referred to the data provided from the internet homepages of Kinema Research & Software (www.kinema.com), JILA (jilawww.colorado.edu), and the data from Hayashi (1987).

The numbers of radicals generated per discharge pulse are calculated by using the method proposed in the previous researches (Rosocha *et al.*, 1993; Mok *et al.*, 1998; Park *et al.*, 1999; Park *et al.*, 2000), and can be expressed as following,

$$X_N = \frac{2k_1}{qv_d(E/N)} \cdot \frac{[N_2]}{N} \cdot \frac{E_p}{V_R} \tag{1}$$

$$X_{N_2A} = \frac{k_2}{qv_d(E/N)} \cdot \frac{[N_2]}{N} \cdot \frac{E_p}{V_R}$$
 (2)

$$X_{O} = \frac{2k_{3} + k_{4}}{qv_{d}(E/N)} \cdot \frac{[O_{2}]}{N} \cdot \frac{E_{p}}{V_{R}}$$

$$\tag{3}$$

$$X_{o^{\dagger}_{D}} = \frac{k_4}{q v_d(E/N)} \cdot \frac{[O_2]}{N} \cdot \frac{E_p}{V_R}$$

$$\tag{4}$$

$$X_{o_2^*} = \frac{k_5}{q v_d(E/N)} \cdot \frac{[O_2]}{N} \cdot \frac{E_p}{V_R}$$
 (5)

where q is the electron charge, E_p the discharge energy per pulse, and V_R the reactor volume. The units of Equations (1)-(5) are number of molecules per discharge pulse. Park *et al.* (2000) shows the calculated results with various reduced electric field (E/N) and that ten times of more oxygen atom (O) is produced than nitrogen atom (N) at typical E/N of 100-200Td (1Td=10⁻¹⁷Vcm²). This higher production of O is due to the lower dissociation energy (5.1 eV) of oxygen than that (9.8 eV) of nitrogen.

The calculated number of radicals generated from electron collision reactions and the measured discharge pulse energy are used as input data to the CHEMKIN-2 software (Kee *et al.*, 1989), which solves the stiff ordinary differential equations to calculate the chemical reactions. 143 species and 791 chemical reactions are

considered for ethene oxidation chemistry. The chemical reactions are based on the kinetic models described in Marinov *et al.* (1998) and Matzing (1991). The kinetic models are crosschecked and modified with the NIST database (Mallard and Mirokhin, 1994) and GRI-Mech3.0 (from internet homepage of GRI-Mech, www.me.berkeley. edu/gri_mech). Ionic reactions in the Matzing's model are not considered in this study since the presence of ionic species does not show any distinctive difference in the chemistry of neutral gas molecules.

It is assumed that there is no appreciable heating of the gas during the plasma reaction, and therefore the reactor is isothermal. The model does not account for axial and radial dispersion in the reactor, i.e., the flow is plug flow. Although the reduced electric field in the reactor varies very rapidly either in time or in space, the field during the pulse period is averaged. The average field of 125Td is assumed by fitting of the kinetic model to the experimental data.

4. RESULTS AND DISCUSSION

4.1. Electric Characteristics of the Discharge

Plots of typical voltage and current waveforms in a mixture of 10% oxygen, base nitrogen at 293K and 473K are shown in Figure 2. Pulsed corona onset (corona initiation voltage) in the reactor is found to occur at about 14.7 kV and 9.1 kV in a mixture of nitrogen and oxygen at 293K and 473K, respectively, and at about 1.6 kV

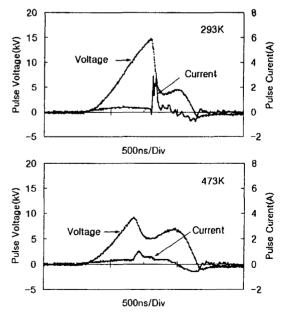


Figure 2. Typical waveforms of pulse voltage and pulse current in a mixture of 10% oxygen, base nitrogen at 293K and 473K.

lower in a mixture of nitrogen only. There is only a slight variation in a corona onset voltage less than ± 0.1 kV for all the test runs. Addition of oxygen between 5% and 20% does not show any noticeable difference in the corona onset voltage with fixed primary input voltage of 136V during the test. The total duration of voltage pulse varies from 1.7 ms to 1.9 ms.

The energy delivered to the discharge pulse is calculated by integrating the products of voltage and current waveforms, and is found to be 4.20 mJ for 293K and 3.02 mJ for 473K. Total energy input to the reactor is controlled by changing the pulse repetition rate from 30 Hz to 140 Hz.

4.2. NO Conversion without Hydrocarbon

Figure 3 shows a comparison of the experiment to the chemical kinetics model for NO to NO_2 conversion in a gas mixture of $10\% O_2$, base N_2 without hydrocarbon addition. As shown in Figure 3, the experimental data are in a fairly good agreement with the calculated data. Even with high energy input, the maximum NO to NO_2 conversion is less than 10%. The main reactions involved in the NO to NO_2 conversion in a gas mixture of nitrogen and oxygen are as following:

$$N+NO \rightarrow N_2+O \tag{R6}$$

$$O+NO+M \rightarrow NO_2+M \tag{R7}$$

$$O+NO_2 \rightarrow NO+O_2$$
 (R8)

$$O+O_2+M \rightarrow O_3+M \tag{R9}$$

$$O_3+NO \rightarrow NO_2+O_2$$
 (R10)

Result of a sensitivity analysis for NO is shown in Figure 4. The result shows that the reactions of (R9) and (R10) play important roles in the NO removal process. In the absence of oxygen, most of NO is reduced to N_2 by reaction (R6) and a very small amount of NO_2 is produced by reaction (R7). However, when oxygen exists in the mixture, the oxidation of NO to NO_2 is accomplished not only by reaction (R7), but also by reactions (R9) and (R10) since ozone production becomes signifi-

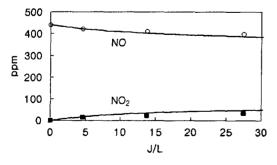


Figure 3. Experimental data (point), compared with the model calculation (line) in a gas mixture of 10% oxygen, base nitrogen with a trace gas of 440 ppm NO at 473K.

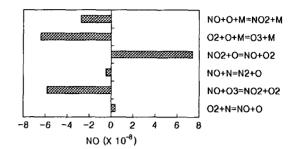


Figure 4. Time integrated sensitivity analysis for NO.

cant with the increase of input energy and the ozone also eventually leads to the oxidation of NO to NO₂. The back conversion of NO₂ to NO via reaction (R8) becomes a limiting factor on the NO to NO₂ conversion efficiency.

4.3. NO Conversion with Ethene

Figure 5 presents the experimental and calculated concentration profiles of NO, NO_2 and ethene in a gas mixture of 10% O_2 , base N_2 with a trace of 250 ppm ethene. The experimental data still shows a good

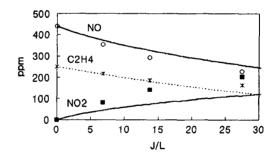


Figure 5. Experimental data (point), compared with the model calculation (line) in a gas mixture of 10% oxygen, base nitrogen with trace gases of 440 ppm NO and 250 ppm ethene at 473K.

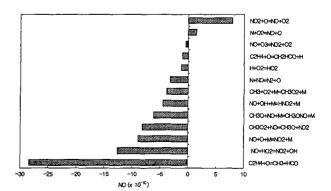


Figure 6. Time integrated sensitivity analysis for NO in the presence of ethene.

agreement with the calculated data, though not so good as in the case of without ethene. The data show that 48% of NO conversion is achieved with the addition of 250 ppm ethene at E/N of 25.4J/L, while it is only 9% without ethene as in Figure 3. The energy required to convert one NO molecule is calculated as 27 eV and 137 eV for with and without ethene, respectively. A sensitivity analysis for this case is shown in Figure 6 and suggests that ozone related reactions (R9) and (R10) do not play important roles any more in NO conversion in the presence of ethene. Instead, oxidation of NO with hydroperoxide (HO₂) and peroxyhydrocarbon (RO₂) becomes very important as in the case of propene addition via following reactions (Penetrante *et al.*, 1997; Park *et al.*, 2000):

$$HO_2+NO \rightarrow NO_2+OH$$
 (R11)

$$RO_2+NO \rightarrow NO_2+RO$$
 (R12)

where R is a hydrocarbon radical. Also the direct oxidizing reaction of NO with O (R7) cannot be neglected.

In the early stage of reaction the ethene reacts with O atom to produce hydrocarbon radicals or intermediates like CH₃, HCO, CH₂HCO:

$$C_2H_4+O \rightarrow CH_3+HCO$$
 (R14)

$$C_2H_4+O \rightarrow CH_2HCO+H$$
 (R15)

$$C_2H_4+O \rightarrow CH_2CO+H_2$$
 (R16)

$$C_2H_4+O \rightarrow C_2H_3+OH \tag{R17}$$

$$C_2H_4+O \rightarrow CH_2O+CH_2$$
 (R18)

The methyl radical, CH_3 , produced from reaction (R14) and the product from the reaction of ethene and OH react with O_2 to form RO_2 , leading to the NO oxidation:

$$CH_3+O_2+M \rightarrow CH_3O_2+M \tag{R19}$$

$$C_2H_4 + OH \rightarrow C_2H_4OH \tag{R20}$$

$$C_2H_4OH+O_2 \to HOC_2H_4O_2 \tag{R21}$$

The HO_2 radicals which convert NO to NO_2 are also produced from the reactions of O_2 with the hydrogen atom and the hydrocarbon intermediates generated from reactions (R14) and (R17), and with the RO generated from reaction (R12):

$$H+O_2 \rightarrow HO_2$$
 (R22)

$$HCO+O_2 \rightarrow CO+HO_2$$
 (R23)

$$C_2H_3+O_2 \to C_2H_2+HO_2$$
 (R24)

$$CH3O+O2 \rightarrow CH2O+HO2$$
 (R25)

During the plasma reaction of ethene, byproducts like formaldehyde (CH_2O), carbon monoxide (CO) and methyl nitrite (CH_3ONO) are produced. Figure 7 shows the calculated concentrations of byproducts when a plasma is applied to a gas mixture of $10\%\ O_2$, base N_2 with 440 ppm NO and 500 ppm ethene at 473K. At a given condition of low energy density, byproduct formation increases with energy density. The model predicts up to 200 ppm of formaldehyde formation at E/N of 23J/L.

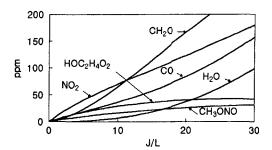


Figure 7. Calculated byproduct formation in a gas mixture of 10% O₂, base N₂ with 440 ppm NO and 500 ppm ethene at 473K.

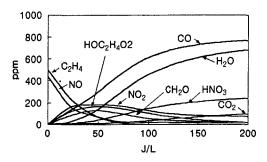


Figure 8. Calculated byproduct formation when enough energy is delivered to the gas mixture as in the same conditions of Figure 7 except 20% O₂.

However, all the ethene and the hydrocarbon intermediates will be partially oxidized to CO and H_2O eventually if enough energy is delivered to the gas mixture as shown in Figure 8.

Figure 9 shows the effect of the initial concentration of ethene on the NO oxidation and the consumption of ethene. The concentrations of NO, NO_2 and ethene are compared by varying the initial ethene concentration from 0-1000 ppm in a gas mixture of $10\%~O_2$, base N_2 at 473K. As the initial ethene concentration increases, the reaction of ethene with O increases, resulting in the promotion of NO conversion.

Figure 10 shows the effect of initial O_2 concentration on the NO oxidation by varying the initial O_2 concentration from 0-20% in a gas mixture of base N_2 with 440 ppm NO and 500 ppm ethene at 473K. As the initial O_2 concentration increases, the production of O by electron collision reactions increases. The increased O and O_2 promote the NO to NO_2 conversion reaction.

Figure 11 shows the effect of gas temperature on the NO oxidation in a gas mixture of 10% O₂, base N₂ with 440 ppm NO and 500 ppm ethene at 273K and 473K. While NO to NO₂ conversion decreases with temperature in the absence of hydrocarbon (Park *et al.*, 2000), it

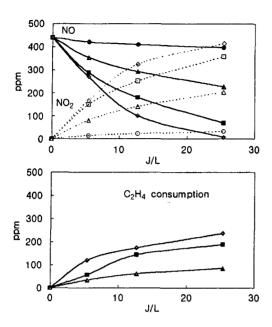


Figure 9. Effect of initial ethene concentration on the NO to NO₂ conversion in a gas mixture of 10% O₂, base N₂ with 440 ppm NO at 473K (\bigcirc : without ethene, \triangle : 250 ppm, \square : 500 ppm, \diamondsuit : 1000 ppm of ethene). Lines are data fits to the experimental data.

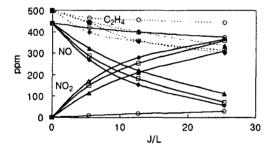


Figure 10. Effect of initial O_2 concentration on the NO to NO_2 conversion in a gas mixture of base N_2 with 440 ppm NO and 500 ppm ethene at 473K (\bigcirc : N_2 only, \triangle : 5%, \square : 10%, \diamondsuit : 20% of O_2). Lines are data fits to the experimental data.

increases with temperature when ethene is added. This is because the reaction rates for reactions (R7) and (R9) that are major reactions in the NO conversion in the absence of hydrocarbon decrease with temperature, while those for reactions of ethene and O increases with temperature.

5. CONCLUSION

The objective of this study is to investigate the effect of ethene on the NO to NO₂ conversion from diesel engine exhaust in a pulsed corona discharge. With the addition

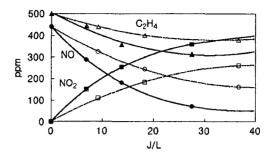


Figure 11. Effect of gas temperature on the NO to NO_2 conversion in a gas mixture of $10\% O_2$, base N_2 with 440 ppm NO and 500 ppm ethene (blank symbol: 273K, filled symbol: 473K). Lines are data fits to the experimental data.

of even a small amount of ethene the NO to NO₂ conversion enhances greatly. The energy required to convert one NO molecule is 27 eV with 250 ppm ethene added, while 137 eV without ethene at 473K. As temperature increases the NO oxidation effect increases since the reaction rates for reactions of ethene and O increases with temperature. Also shown are the effects of the initial concentrations of ethene and oxygen, both of which promote the reaction of ethene and O radical, resulting in the increase of NO conversion rate.

A kinetic model was developed to characterize the NO oxidation process during corona discharge. The model predictions show good agreement with the experimental data. Byproducts like formaldehyde (CH_2O) and methyl nitrite (CH_3ONO) are predicted by model calculation, but both of them are broken up into CO and H_2O eventually when high energy is delivered to the gas mixture. Sensitivity analysis shows that the main pathways of NO oxidation when ethene is added are: where R is a hydrocarbon radical. But, the direct oxidizing reaction of NO with O cannot be neglected.

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