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Reactive Flow Analysis of Nonthermal Plasma in a Cylindrical Reactor

Takehiko Sato, Member, IEEE, Dai Ito, and Hideya Nishiyama

Abstract—In the study reported herein, chemical reactive flow fields of nonthermal plasma were clarified by using both experimental and numerical methods. The plasma was generated by using needle-plate electrodes. The operating gas was air injected at a rate of 1–10 NL/min through a cylindrical reactor with a diameter of 6 mm. Applied voltage of 12 kV was positive or negative with a square wave of 20 Hz. Distributions of gas velocity, gas temperature, and NO and NO$_2$ concentrations were analyzed by experimental and numerical methods. Conservation equations of mass, chemical species, momentum and energy, and 22 chemical species and 89 chemical reactions are taken into consideration in the model herein proposed. Distributions of axial velocity and gas temperature, their concentrations of NO and NO$_2$, and the effects of gas flow rate were clarified. For these characteristics of the parameter, the same tendency was obtained by both experimental and numerical methods. Furthermore, distributions of radical species such as N and O were predicted by numerical analysis using experimental results.

Index Terms—Air, atmospheric pressure, computational experiment, nonthermal plasma, radical, reaction.

I. INTRODUCTION

Air pollution by flue gas, exhaust gas, and volatile organic compounds (VOCs) causes acid rain and global warming. Chlorofluorocarbons (CFCs), well-known VOCs, have a significant greenhouse effect. Furthermore, our living space, including houses and offices, is polluted by VOCs emitted by new construction material, wall paint and wallpaper, and carpet adhesives. VOC pollution in our living space has become a serious social problem because it causes sick house and building syndrome.
Nonthermal plasma generated by high-voltage discharge at atmospheric pressure has been recently utilized for environmental applications such as ozone generation for sterilization, fossil fuel conversion, and decomposition of VOCs, NO\textsubscript{x} and SO\textsubscript{2}. In particular, decomposition of low-concentration VOCs using nonthermal plasma flow has gained attention as a mean of preventing such syndrome, because it provides high chemical reactivity and a proper decomposition process even for low concentration VOCs.

However, the plasma decomposition system is not yet well commercialized because there are still unknown chemical processes involved in its complex chemical reactions, although its discharge processes [9]–[12] and the gas flow effect on the breakdown voltage [13] have been investigated.

The objective of the present study is to clarify the chemical reaction fields of a nonthermal plasma flow using both experimental and numerical methods to gain fundamental information on the chemical process. It is very important to clarify the reactive flow field using only air in the downstream of the discharge part, because the radical species and by-products, i.e., NO\textsubscript{2} and so on, which contribute to the chemical process in applications such as VOCs decomposition, are mainly generated from air components such as oxygen, nitrogen and water. We focused on NO\textsubscript{x} concentrations, which can be measured experimentally, to estimate the radical species distribution. The data and the method obtained in this study would help very much to solve the chemical process in the nonthermal plasma such as VOCs decomposition process.

II. EXPERIMENTAL ANALYSIS

Fig. 1 shows a schematic of an experimental apparatus. It consists mainly of a function generator, an amplifier, a cylindrical reactor, a gas supply system and a measurement system. Corona discharge is generated by using needle-plate electrodes, the gap length of which is 1 or 2 mm at atmospheric pressure. Operating gas is air injected at a rate of 1–10 NL/min through a cylindrical reactor. Applied voltage of 12 kV is positive or negative with a square wave of 20 Hz and the pulse rise time is within 24 \mu s as shown in Fig. 2.

Fig. 3 shows cross sections of the cylindrical reactor, which is made of Teflon. Its diameter at the discharge part is 6 mm.

A numerical model is based on the following assumptions: 1) electrons and ions are not considered in the nonthermal plasma; 2) the flow is continuous, incompressible, and laminar; 3) the flow, temperature, and concentration fields are two-dimensionally axisymmetric; 4) gravity is negligible; and 5) radiation loss and absorption are not considered.

B. Governing Equations

Under the above-mentioned assumptions, the governing equations are presented as follows [14].

Continuity equation
\[ \nabla \cdot \mathbf{u} = 0. \]  

Momentum equation
\[ \rho(\mathbf{u} \cdot \nabla)\mathbf{u} = -\nabla p + \mu \nabla^2 \mathbf{u}. \]

Energy conservation
\[ \rho \nabla \cdot (h \mathbf{u}) = \lambda \nabla^2 T + \Phi_D + S_C. \]

Transport equation for chemical species, \( i \)
\[ \rho \nabla \cdot (Y_i \mathbf{u}) = \nabla \cdot \mathbf{J}_i + \omega_i. \]
Here, \( J_i \) and \( \omega_i \) are defined as follows:
\[
J_i = - \rho D_i \nabla Y_i \tag{5}
\]
\[
\omega_i = W_i \sum_{k=1}^{K} k_{ij} (v'_{ij} - u'_{ij}) \prod_{j=1}^{N} c^{l'_{ij}}. \tag{6}
\]
Here, the \( k_{ij} \) is reaction rate defined as \( k_{ij} = A_k T^n \exp\left(-E_k/RT\right) \). Twenty-two chemical species and 88 chemical reactions are taken into consideration in this model [15]. The main chemical reactions are shown in Table I.

### C. Computational Domain and Boundary Conditions

Fig. 4 shows a computational domain. The domain is adopted downstream from the electrode position with the same configuration as that of the experimental reactor. The boundary conditions are given as follows:

- **Intel**: \( u = u_0, v = 0, T = T_0, Y_i = \text{given} \)
- **Axis**: \( \frac{\partial u}{\partial r} = v = 0, \frac{\partial T}{\partial r} = 0, \frac{\partial Y_i}{\partial r} = 0 \)
- **Wall**: \( u = v = 0, T = \text{const.}, \frac{\partial u}{\partial n} = 0 \)
- **Exit**: \( \frac{\partial T}{\partial x} = \frac{\partial u}{\partial x} = \frac{\partial Y_i}{\partial x} = 0 \)

Here, \( n \) is the direction normal to the wall.

### IV. RESULTS AND DISCUSSION

Symbols and lines in the figures show experimental results and numerical results, respectively.

Fig. 5 shows the radial distribution of the axial velocity of a gas flow. When the gas flow rate is lower than 3 NL/min, the parabolic radial distribution of axial velocity at the exit of the cylindrical reactor shows good agreement between the experimental and numerical results. The flow is laminar as the Reynolds number is lower than 1800. The radial distribution of the measured axial velocity is trapeziform when gas flow rate is higher than 6 NL/min, because the turbulent flow is developed due to a high Reynolds number of 5300. On the other hand, the radical distribution of the simulated axial velocity is not fully developed when the gas flow rate is higher than 6 NL/min, because a uniform velocity at the inlet is given.

Fig. 6 shows the axial distribution of gas temperature in a cylindrical reactor. The gas temperature increases under conditions of lower gas flow rate due to the higher enthalpy.

### TABLE I

<table>
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<tr>
<th>Chemical reaction</th>
<th>Reaction rate (( \text{cm}^2/\text{sec} ))</th>
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<tr>
<td>( \text{O}_2 + \text{NO}_2 \rightarrow \text{O}_3 + \text{NO} )</td>
<td>( 6.4 \times 10^{-8} \exp(663/T) )</td>
</tr>
<tr>
<td>( \text{O}_2 + \text{O}_3 \rightarrow 2 \text{O}_2 )</td>
<td>( 1.8 \times 10^{-11} \exp(2300/T) )</td>
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<tr>
<td>( \text{NO}_2 + \text{H}_2 \rightarrow \text{NO} + \text{H}_2 )</td>
<td>( 5.7 \times 10^{-34} \exp(2200/T) )</td>
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<tr>
<td>( \text{NO} + \text{H}_2 \rightarrow \text{NO} + \text{H}_2 )</td>
<td>( 2.4 \times 10^{-11} \exp(4000/T) )</td>
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<tr>
<td>( \text{NO} + \text{H}_2 \rightarrow \text{NO} + \text{H}_2 )</td>
<td>( 3.1 \times 10^{-11} \exp(1200/T) )</td>
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<tr>
<td>( \text{NO} + \text{H}_2 \rightarrow \text{NO} + \text{H}_2 )</td>
<td>( 6.0 \times 10^{-12} )</td>
</tr>
<tr>
<td>( \text{NO} + \text{H}_2 \rightarrow \text{NO} + \text{H}_2 )</td>
<td>( 2.0 \times 10^{-12} )</td>
</tr>
<tr>
<td>( \text{NO} + \text{H}_2 \rightarrow \text{NO} + \text{H}_2 )</td>
<td>( 9.5 \times 10^{-33} \exp(1300/T) )</td>
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<tr>
<td>( \text{NO} + \text{H}_2 \rightarrow \text{NO} + \text{H}_2 )</td>
<td>( 8.6 \times 10^{-32} \exp(1300/T) )</td>
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<tr>
<td>( \text{NO} + \text{H}_2 \rightarrow \text{NO} + \text{H}_2 )</td>
<td>( 1.0 \times 10^{-31} \exp(1300/T) )</td>
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<tr>
<td>( \text{O} + \text{H}_2 \rightarrow \text{OH} + \text{H} )</td>
<td>( 9.0 \times 10^{-18} )</td>
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<tr>
<td>( \text{O} + \text{H}_2 \rightarrow \text{OH} + \text{H} )</td>
<td>( 2.3 \times 10^{-11} \exp(1000/T) )</td>
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### TABLE II

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<td>Inlet velocity (m/s)</td>
<td>0.67</td>
<td>1.37</td>
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<td>4.92</td>
<td>7.5</td>
<td>4.92</td>
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<tr>
<td>Inlet temperature (K)</td>
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<td>287</td>
<td>285</td>
<td>284</td>
<td>283</td>
<td>284</td>
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<tr>
<td>Inlet chemical species compositions (wt %)</td>
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<td></td>
<td></td>
<td></td>
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<tr>
<td>O</td>
<td>( 1 \times 10^{-2} )</td>
<td>( 1 \times 10^{-4} )</td>
<td>( 3.3 \times 10^{-4} )</td>
<td>( 2.0 \times 10^{-4} )</td>
<td></td>
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</tr>
<tr>
<td>H</td>
<td>( 1 \times 10^{-7} )</td>
<td>( 3.3 \times 10^{-8} )</td>
<td>( 2.0 \times 10^{-8} )</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>( \text{H}_2 )</td>
<td>( 1 \times 10^{-7} )</td>
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<tr>
<td>( \text{H}_2 )</td>
<td>( 1 \times 10^{-7} )</td>
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D. Computational Conditions

Table II shows computational conditions. Inlet velocity, inlet temperature, and ambient temperature are given by using experimental data. Compositions of the chemical species at the inlet are determined by adjusting the inlet conditions to obtain good agreement between the axial distributions of experimental and numerical data, because the axial distribution of NO, one of the end products, is easily measured experimentally, and because the axial distribution would show the basic characteristics of the chemical process. Therefore, the axial distribution is very important to predict more accurate radical species distribution.

The velocity, temperature and concentration fields are simulated by using CFD-ACE+ code (CFD Research Corporation, Huntsville, AL) which employs the SIMPLEC method.
gas temperature decreases steeply in the adjacent discharge region. While the same tendency is shown by numerical analysis, the gas temperature is the highest in case 3 compared with other cases around $x = 10$ mm. On the other hand, the lowest temperature is obtained in case 3 downstream of the point of $x = 30$ mm.

Fig. 7 shows the axial distribution of NO$_2$ concentration. When the gas flow rate is 2 NL/min, NO$_2$ concentration increases steeply initially and then shows the maximum around $x = 100$ mm. However, it increases gradually when the gas flow rate is higher than 6 NL/min. The quantitative agreement between experimental and numerical results are shown here.

Figs. 8 and 9 show, respectively, NO and NO concentrations obtained numerically in case 2 ($Q = 2$ NL/min). The distributions of NO$_2$ and NO concentrations are influenced by the flow velocity distribution because convective transportation distance depends on the flow velocity. NO$_2$ concentration strongly depends on the NO concentration. NO is generated in the vicinity of the inlet and shows the maximum value there. Correspondingly, NO$_2$ increases with decreasing NO concentration as shown in Table I.

Fig. 10(a) and (b) shows O and N concentrations, respectively, in case 2. Fig. 11(a) and (b) shows O and N concentrations, respectively, in case 5. O and N show high concentrations at the inlet and decrease steeply with the increase in the $x$ direction until 2 mm in case 2 due to low velocity. However, N concentration is widely diffused downstream in case 5 due to high velocity, because the chemical reaction rate of N is slower than that of O. This causes the lower increase of NO and this process finally causes a slow increase of NO$_2$ concentration at the high gas flow rate. Therefore, one major factor contributing to NO$_2$ generation is N distribution.

Fig. 12 shows the axial distribution of radical species concentrations in case 5. The O$_3$ which is important to generate and disappear NO and NO$_2$, are increased rapidly near the inlet. The
Fig. 10. (a) O and (b) N concentrations in case 2.

Fig. 11. (a) O and (b) N concentrations in case 5’.

Fig. 12. Axial distribution of radical species concentrations in case 5’.

OH is decreased with increase in the NO due to react with N. These results imply that the computational experiment using experimental data is an effective method to understand the complex reactive flow.

V. CONCLUSION

In the present study, chemical reaction flow fields of non-thermal plasma were clarified by using both experimental and numerical methods to gain fundamental information on decomposition process of VOCs. The results obtained can be summarized as follows:

1) The parabolic radial distributions of axial velocity show good agreement between the experimental and numerical analyses.
2) Axial distributions of gas temperature decrease steeply when the gas flow rate is low, which is shown by both experimental and numerical analyses.
3) NO\textsubscript{2} concentration strongly depends on the NO concentration. One major factor contributing to NO\textsubscript{2} generation is, therefore, N distribution.
4) It is possible to predict the radical species, which cannot be easily measured, by integrating experimental and numerical analyses.

REFERENCES


Takehiko Sato (M’05) received the Bachelor’s, Master’s, and Ph.D. degrees in mechanical engineering from Tohoku University, Sendai, Japan, in 1990, 1992, and 1995, respectively.

In 1995, he joined Matsushita Electric Works, Ltd. In 1998, he joined the Institute of Fluid Science, Tohoku University, where he is currently an Associate Professor. His research interests have included control of a nonequilibrium plasma flow and numerical evaluation of a plasma spraying process. Recently, his efforts have been concentrated on treatment of indoor environments using a low-temperature plasma flow at atmospheric pressure.

Dai Ito received the Bachelor’s and Master’s degrees in mechanical engineering from Tohoku University, Sendai, Japan, in 2001 and 2003, respectively. In 2003, he joined NSK Ltd., Tokyo, Japan.

Hideya Nishiyama received the Bachelor’s, Master’s, and Ph.D. degrees in mechanical engineering from Tohoku University, Sendai, Japan, in 1977, 1979, and 1982, respectively.

He has been a Professor since 1997 and the University Councillor since 2004 at the Institute of Fluid Science, Tohoku University. He was a Research Associate in 1982, a Lecturer in 1985, and an Associate Professor in 1988 at the Mining College, Akita University, Japan. He was an Associate Professor at the Institute of High Speed Mechanics, Tohoku University, in 1989. He was a Visiting Researcher at the Von Karman Institute for Fluid Dynamics, Belgium.

Prof. Nishiyama has been awarded the Excellent Prize from the Institute of Theoretical and Applied Mechanics, Siberian Branch of the Russian Academy of Sciences, the Japan Society of Mechanical Engineers Best Paper Award and the Japanese Society for Multiphase Flow Best Paper Prize.