Compatibility of Fuels and Radicals Found in Plasma Jets for Improved Premixed Combustion

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(Received October 9th, 2010)

We examined the compatibility of radicals contained in plasma jets to fuels through ignition and combustion tests for dimethyl ether (DME)/air and methane (CH₄)/air mixtures with oxygen (O₂) and nitrogen (N₂) as the plasma torch feedstocks. The experiment showed that the DME/air mixture was ignited/combusted with less plasma jet (P.J.) power than the CH₄/air mixture and that the O₂ P.J. is more effective than the N₂ P.J., with a more distinct difference in effectiveness for the CH₄/air mixture in contrast to the DME/air mixture. Plasma jets with fewer feedstock flow rates were more effective, presumably due to the greater amount of radical production under the conditions tested. Numerical estimation on the amount of radicals and ignition delay time demonstrates that the superiority of the O₂ P.J. is not necessarily only due to the effectiveness of the O radicals, but also due to the fact they were produced easier and with less power, and that the effect and behavior according to amount is different for fuels. This is most likely because they depend on the reaction mechanism of each mixture, all of which match well with the experimental results.

Key Words: Combustion, Plasma Jets, Radicals, DME, Methane

Nomenclature

\[ P_{in} \] : input electric power
\[ P_{c,min} \] : minimum power for combustion
\[ P_{i,min} \] : minimum power for ignition
\[ Q_{r} \] : flow rate of plasma jet feedstock
\[ V_{in} \] : premixed gas flow speed
\[ \phi \] : equivalence ratio of premixed gas
\[ t_{ig} \] : ignition delay time
\[ Qr \] : produced radical rate

1. Introduction

Various research relating to the effects of radicals on ignition and combustion have been conducted1-5) because radicals are understood to play important roles in chemical reactions. However, most research is for cases with specific situations, such as a given flow condition or combustor configuration. A method to evaluate a more general effect of each radical on combustion reaction would be a vital and useful tool for developing higher-performance or innovative combustion systems, including the use of new alternative fuels.

Accordingly, we have proposed and tried to develop a new combustion experimental apparatus that allows radicals to be produced onsite and implanted in a premixed gas. A combustion test setting coupled with a plasma torch with appropriate feedstock for producing and adding radicals to mixture gases was designed, and has been used for a series of studies.

In a previous report,6) we showed that the proposed approach was appropriate for evaluating and comparing the effectiveness of each radical species in terms of ignition limits and combustion completeness for dimethyl ether (DME), CH₃OCH₃, a promising next-generation alternative fuel,7) as a test trial case under some conditions.

In the current report, more experiments were conducted under additional conditions with another very common gas fuel, methane (CH₄). The results are intended to be used to clarify the congeniality of each radical kind found in plasma jets to the kind of fuel tested from the viewpoint of promoting ignition and combustion.

2. Experimental Apparatus and Test Procedure

With the experimental set-up, a schematic diagram of which is shown in Fig. 1, the test procedure is as follows: Premixed gas at a set flow rate and mixture ratio is supplied into the combustion chamber where air is used after being dried as an oxidizer, and DME and CH₄ are used as fuel. Then, a plasma jet with a test feedstock (oxygen (O₂) or nitrogen (N₂) in this study) is initiated and operated at a given power. High-temperature plasma jets (P.J.) including many active radicals are spread and supplied to premixed gases flowing coaxially, there by igniting them chemically and thermally.

While ejecting plasma jets into fuel/air mixtures, the
electric current/voltage, temperature and carbon dioxide (CO₂) concentration are measured at 85 mm and 160 mm downstream from the P.J. exit, respectively. Incidentally, water in the exhaust is excluded in the CO₂ concentration account. Completeness of combustion is judged from CO₂ concentration and temperature, as well as by visual observation in the chamber for each P.J. input power, P_{in}. CO₂ concentration and temperature without fuel are also measured at the same airflow and P.J. condition to ensure the influence of plasma jets on the temperature is negligible. The diameter of the P.J. torch nozzle is 1 mm.

**Fig. 1.** Schematic diagram of the experimental set-up.

### 3. Results and Discussion

#### 3.1. Effect of feedstock type

Ignition and combustion tests with oxygen plasma jets (O₂ P.J.) and nitrogen plasma jets (N₂ P.J.) were conducted in order to compare the effectiveness of oxygen atoms and nitrogen atoms in DME-air mixture combustion; spectroscopic measurements in a previous paper⁹ reported the presence of O and N atoms in O₂ P.J. and N₂ P.J., respectively.

Figure 2 shows the relation between CO₂ concentration, temperature increase and input electric power P_{in} for DME/air combustion at the equivalence ratio φ = 0.6 using O₂ P.J. and N₂ P.J. The experiments were performed under atmospheric pressure and room temperature conditions at a plasma jet feedstock flow rate of Q_{f} = 2 NL/min and premixed gas flow speed of V_{m} = 9 m/s. Judgment of ignition/non-ignition and combustion state was made using CO₂ concentration trends coupled with the temperature increase shown in Fig. 2. The horizontal chain lines in the figures indicate CO₂ concentration in the exhaust gas, except for the water calculated by a chemical equilibrium program (CEA)⁸ for the combustion of a DME-air mixture at φ = 0.6. As shown in Figs. 2(a)-2(b), there are three distinct zones of CO₂ concentration: Zone 1, where CO₂ concentration hardly increases further even if input power is increased; Zone 2,

**Fig. 2.** Relation between CO₂ concentration, temperature increase and input electric power for DME/air combustion at φ = 0.6 using oxygen and nitrogen plasma jets.
where it sharply increases as input power increases; and Zone 3, where no CO2 is detected.

Together, these indicate that combustion of the DME/air mixture is not observed in Zone 3, is partially complete in Zone 2, and is more complete toward Zone 1. The vertical broken lines in the figures indicate \( P_{i,min} \) and \( P_{t,min} \), which separate Zones 2 and 3 and Zones 1 and 2, respectively. Though the minimum power necessary for ignition \( P_{i,min} \) does not differ much for each plasma jet, the oxygen plasma jet requires somewhat less power for combustion \( P_{t,min} \). Therefore, O2 P.J. is slightly more effective than N2 P.J. for promoting combustion of the DME/air mixture.

### 3.2. Effect of feedstock flow rate

Figure 3 shows the relation between CO2 concentration, temperature increase and input electric power for DME/air combustion at \( \phi = 0.6 \). The oxygen plasma jets were operated at \( Q_j = 1 \) NL/min and 3 NL/min to examine the effect of the P.J. feedstock flow rate. Comparing Figs. 3(a)-3(b) to Fig. 2(a), \( P_{i,min} \) and \( P_{t,min} \) are the lowest at \( Q_j = 1 \) NL/min, and hardly differ in the cases of \( Q_j = 2 \) and 3 NL/min. The results indicate that the lower the flow rate of the plasma jet feedstock is, the more effectively active species are produced and emitted in the range of \( P_{in} \) and \( Q_j \) in this study.

### 3.3. Effect of fuel type

Figure 4 shows the relation between CO2 concentration, temperature increase and input electric power for CH4/air combustion at \( \phi = 0.6 \) to examine the effect of the type of fuel used. The experiments were performed under atmospheric pressure and room temperature conditions at \( Q_j = 2 \) NL/min and \( V_m = 9 \) m/s. The overall experimental results are summarized in Table 1. These indicate that \( P_{i,min} \) and \( P_{t,min} \) for DME/air are lower than those for CH4/air (which means DME is easier to use for ignition and combustion), and that those using O2 P.J. are lower than those using N2 P.J. for both DME/air and CH4/air, whereas the difference in the effectiveness between O2 P.J. and N2 P.J. is more distinct for the CH4/air. The findings for the CH4/air mixture agree with the reaction promotion effect of oxygen atoms obtained in past studies.2, 41

### 3.4. Discussion using numerical simulation

In order to examine the experimental results, numerical evaluations were estimated. Figure 5 shows temperature and the amount of O and N atoms produced in O2 and N2 P.J. in the range of \( P_{in} \) used in this study as calculated by CEA. The amount of radicals is expressed by volumetric percent in the total main flow (\( V_m = 9 \) m/s or equivalently 20.8 NL/min). Although it is understood that the real temperature and radical densities are not the averaged ones shown here because of their gradients in the jets,2) such information on their behavior with \( P_{in} \) is somewhat useful. In addition, Fig. 6 shows the ignition delay and radical flow rates in the case of O2 P.J. for DME/air at \( \phi = 0.6 \), the initial temperature of 1300 K calculated based on Fig. 5. The radical amount in the right ordinate in the graph is expressed by the volumetric flow rate to compare with the total main flow, 20.8 NL/min. Not only O atoms in O2 P.J., but also non-dissociated O2 are included for the

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**Fig. 3.** Relation between CO2 concentration, temperature increase and input electric power for DME/air combustion at \( \phi = 0.6 \).
calculation of ignition delay. Ignition delay is defined as the time required for a rise of 400K from the initial temperature. The DME/air oxidation reaction used for the calculation here is based on a mechanism\(^9\) combining Kaiser’s mechanism\(^10\) with modifications to the Miller-Bowman mechanism, which treats the nitrogen-oxide mechanisms of C\(_1\)-, C\(_2\)-hydrocarbon fuels.\(^11\) In the range of the DME/air flow rate and O\(_2\) P.J. operating conditions in this study, ignition delay is the least at 1 NL/min among the three O\(_2\) P.J. flow rates, and is about the same for the cases of 2 and 3 NL/min.

As shown in Fig. 5, up to \(P_{in} = 100\) W temperatures of nitrogen and oxygen increase at almost the same rate and no dissociation appears. Over 100 W, the temperature difference between N\(_2\) P.J. and O\(_2\) P.J. becomes wider. This is due to the appearance of larger dissociation of O\(_2\) to O earlier, at lower \(P_{in}\) or lower temperatures, than that of N\(_2\) to N; distinct dissociation starts at 100 W for O\(_2\), but 200 W for N\(_2\). Once dissociation starts, temperature increase fails. A larger amount of O atoms in the plasma jets is more effective than raising the temperature of the N\(_2\) plasma jets, since it was observed that O\(_2\) P.J. was more effective for promoting combustion at the same input electric power.

Figure 7 shows a calculated ignition delay time for each plasma jet with \(Q_i = 2\) NL/min, for each fuel mixture at \(\phi = 0.6\), initial temperature 1300 K, and \(V_a = 9\) m/s. GRI-Mech 3.0\(^12\) was used for the CH\(_4\)-air mixture oxidation reaction mechanism.

Ignition delay time around \(P_{in} = 100\) W is the shortest for DME/air with O\(_2\) P.J., followed by DME/air using a N\(_2\) P.J., CH\(_4\)/air with O\(_2\) P.J., and CH\(_4\)/air with N\(_2\)P.J., which agrees with the tendency of experimental results for \(P_{in}\) and \(P_{c,min}\). The experimental result showing that O\(_2\) P.J. is more effective than N\(_2\) P.J. would be explained by the numerical results where O atoms by O\(_2\) P.J. are produced more efficiently than N atoms by N\(_2\) P.J., as is shown in Figs. 5 and 6. As for the ignition delay CH\(_4\)/air with O\(_2\) P.J., shortened steeply over \(P_{in} = 100\) W to be the shortest as shown in Fig. 7, it is presumed to be due to the fact that O atoms behave more effectively for promoting reaction in CH\(_4\)/air than in DME/air in that region. These numerical results offer a good explanation in qualitative terms the experimental results, such as those shown in Table 1.

### Table 1. Marginal input electric power necessary for \(P_{c,min}\) and \(P_{i,min}\)

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Feedstock</th>
<th>(P_{c,min}) (W)</th>
<th>(P_{i,min}) (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DME</td>
<td>(O_2)</td>
<td>80</td>
<td>140</td>
</tr>
<tr>
<td></td>
<td>(N_2)</td>
<td>80</td>
<td>150</td>
</tr>
<tr>
<td>CH(_4)</td>
<td>(O_2)</td>
<td>90</td>
<td>165</td>
</tr>
<tr>
<td></td>
<td>(N_2)</td>
<td>100</td>
<td>195</td>
</tr>
</tbody>
</table>

Fig. 4. Relation between CO\(_2\) concentration, temperature increase and input electric power for CH\(_4\)/air combustion at \(\phi = 0.6\).
4. Conclusion

Ignition and combustion tests using an experimental apparatus with a plasma torch were carried out to evaluate the effectiveness of the radicals contained in the plasma jet for DME/air and CH$_4$/air mixtures. The results showed:

1) DME/air mixture was ignited and burned more easily when using plasma jets; that is, with less P.J. power than the CH$_4$/air mixture.

2) O$_2$ P.J. is more effective than N$_2$ P.J. for promoting combustion, with a more distinct difference for the CH$_4$/air mixture than for the DME/air mixture.

3) Plasma jets with lower feedstock flow rates showed higher effectiveness because more active atoms were produced and emitted in the range of the flow rates and $P_{in}$ tested in this study.

Numerical simulations conducted to examine the above experimental results indicate:

4) With each amount of O and N atoms estimated to be produced by the same P.J. input power $P_{in}$, having additional O atoms is superior to that of N atoms for shortening the ignition delay. In particular, the difference in effect is more distinct in CH$_4$/air than in DME/air, the tendency of which seems to be demonstrated by the experimental results on $P_{c,min}$ as well.

The experiment and numerical estimations in this study suggest that combustion processes are affected complexly by the type of radical and its density in P.J. with the local
temperature near the reaction area. Decoupling the temperature effect from the radical effect itself is indisputably an attractive topic for further research.

References


