Ignition of CH$_4$ by Counterflowing Hot Air Diluted with Combustion Products

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Abstract
Ignition temperature of CH$_4$ by counterflowing hot air diluted with combustion products was investigated numerically using a flame-controlling continuation. GRI-v3.0 was used for the calculation of chemical reaction rates in all the computations. The maximum H radical concentrations were presented in the function of the global strain rate, and showed S-curve behavior. Ignition temperature of CH$_4$ was determined using S-curve of the H radical concentration. The effects of strain rate on the ignition temperature of CH$_4$ were investigated with the dilution rate ($\Omega$) of combustion products. Pollutant emission characteristics and flame structures were investigated with a steady counterflow flame code, OPPDIF. The emission indices for major pollutant were introduced to compare the pollutant emission quantitatively. EINOx for the condition considered in this study ($\Omega \leq 0.5$) was still higher than that of usual low-temperature air nonpremixed combustion.

Introduction
The global warming and environmental pollution problems arise from the combustion systems that utilize the fossil fuels becomes more serious recently. Combustion process of the fossil fuels essentially emits green-house gases and pollutant, such as NOx, SOx, and soot. Thus, every effort to reduce such green-house gas and pollutants is progressing all over the world. The best way to reduce the main green-house gas, CO$_2$, emitted during the combustion of fossil fuels is to enhance the combustion efficiency. High-temperature air combustion is a promising technique to improve the combustion efficiency as well as to reduce the pollutant emission. High-temperature air combustion improves the combustion efficiency by recycling exhaust heat to the supplying air stream [1, 2]. In addition, it was known that flameless combustion with small temperature gradient in all direction can be achieved by making the supplying air stream be high-temperature and low-oxygen content oxidizer. This flameless combustion can be possible when the temperature of supplying air stream is over the auto-ignition temperature of fuel. The oxygen concentration in air stream is closely related with the pollutant emission from the flameless combustion.

It has been known that there are three combustion regions in the high-temperature air combustion in the respect of mixing of fuel and air [1]. The first one is BF/BA region where the combustion reactions occur between diluted fuel with burnt gases (BF) and diluted air with burnt gases (BA). Reactions between diluted fuel with burnt gases and fresh air (A) occur at the second region (BF/A), and combustion between fresh fuel (F) and diluted air with burnt gases at the third region (F/BA). Previous fundamental studies regarding the high-temperature air combustion have mainly focused on the BF/A region [3,4]. It is necessary to investigate the combustion characteristics in the other regions, such as F/BA and BF/BA, to understand the high-temperature air combustion in more detail.

Thus, the main objective of this paper is to investigate the auto-ignition temperature of CH$_4$ by counterflowing hot air diluted with combustion products for considering the high-
temperature air combustion characteristics in F/BA region. In addition, flame structure and pollutant emission characteristics of the F/BA combustion region were investigated.

**Numerical Procedure**

An OPPDIF code [5] usually used for the simulation of steady counterflow flame is not applicable for the investigation of ignition since which is unsteady phenomenon essentially. In this study, a flame-controlling continuation method (FCCM) developed by Nishioka et al. [6] was used to obtain an ignition temperature of counterflowing CH\(_4\) by hot air mixed with combustion products. The maximum H concentration was presented in the function of the inverse global strain rate, and showed S-curve behavior related with the extinction and ignition. The numerical algorithm of FCCM is very similar to that of OPPDIF except boundary conditions and dependent variables. The detailed numerical procedure of FCCM was described elsewhere [6]. The OPPDIF code was used for steady counterflow flame structure and pollutant emission characteristics. Steady OPPDIF results were used as initial conditions for the computation with FCCM.

In this study, we focus on the ignition and emission characteristics of F/BA region among the highly-preheated air combustion regions. Thus, the air stream was mixed with the combustion products and its temperature was raised sufficiently such that inflowing fuel was ignited. The concentration of the combustion products was computed using UPSR (Unsteady Perfectly-Stirred Reactor) code [7]. The UPSR code computed the temperature and species concentration for the condition where fuel and air are perfectly mixed. First, the concentrations of main combustion products of CH\(_4\) were evaluated for a condition of the equivalence ratio (Φ) of 1.0 using UPSR. All species concentrations formed from the pyrolysis of the main combustion products and air at supplying high-temperature air stream were also computed with UPSR. The concentrations of all the species were used as the inflow boundary condition when steady flame structure was computed using OPPDIF code.

**Numerical Conditions**

In order to investigate the ignition characteristics of fuel, flow field that consisted of counterflowing CH\(_4\) and hot air streams was used as a geometry in this study. The separation distance (L) between fuel and air nozzles was 15 mm. For chemical reactions GRI-v3.0, which considers 53 species and 325 reactions, was used for all computations. The radiation effects were not considered in this study.

The temperature of fuel stream was fixed to 300 K, and the temperature of air stream mixed with the combustion products was varied from lower value to sufficiently-higher one. The pressure was fixed to 1 atm for all conditions. The volume fraction of O\(_2\) and N\(_2\) was 21\% and 79\%, respectively, for pure air stream. In case that the combustion products were mixed with pure air, concentrations of the combustion products were varied to the dilution rate (Ω). The dilution rate was defined as follows:

\[
Ω = \frac{\text{volume of product gases}}{\text{volume of total gases in air stream}}
\]  

(1)

A global strain rate (\(a_g\)) [8], which is important parameter to characterize diffusion flames, was introduced in this study.

\[
a_g = \frac{2V_a}{L} \left( 1 + \frac{V_f \sqrt{\rho_f}}{V_a \sqrt{\rho_a}} \right)
\]  

(2)
where, $V_f$ and $V_a$ is separately velocity of fuel and oxidizer, and $\rho_f$ and $\rho_a$ is separately density of fuel and oxidizer.

In order to quantify the emission characteristics of pollutants, emission indices [9, 10] for CO, CO$_2$, NO and NO$_2$ were evaluated using the following equation (3):

$$EI_i = \frac{\int_0^L W_i \dot{\omega}_i dx}{-\int_0^L W_{CH4} \dot{\omega}_{CH4} dx}$$

(3)

where, $i$ is the species CO, CO$_2$, NO and NO$_2$; $W_i$ is the molecular weight of species $i$; $\dot{\omega}_i$ is the molar production rate of species $i$; $L$ is the separation distance between fuel and oxidizer nozzles.

**Results and Discussion**

It was known that the maximum H radical concentration versus strain rate provides more precise ignition temperature data than the maximum temperature [6, 11]. In this study, thus, we adopted the maximum H radical variation for the investigation of ignition temperature. Fig. 1 shows the maximum H radical concentration versus the inverse of the global strain rate for case of pure air stream. For investigation of ignition characteristics, the temperature of inflowing air stream ($T_O$) was varied from 700 K to 1270 K. The variation of the H radical shows a typical S-curve behavior that is directly related with the extinction and ignition characteristics of nonpremixed flame of CH$_4$-pure air. The first turning points located at higher concentration of H radical are extinction points for each inflowing condition, and the second turning points located at lower concentration are ignition points. For $T_O = 700$ K and 900 K, the ignition points were not identified for considered strain rate range. This means that the fuel cannot be ignited for the strain rate range considered. At $T_O \geq 1100$ K, the shapes of second turning points were not monotonous but two-stage behavior although the second turning points existed apparently. For case of two-stage-ignition, $I_1$ and $I_2$ were known as primary and secondary ignition points, respectively.

Fig. 2 shows the maximum H radical response to the global strain rate when the combustion products are mixed in the air stream. The dilution rate, $\Omega$, was varied to 0 (pure air) to 0.5. At $\Omega \geq 0.4$, the second turning point was not observed. In previous studies [12-16], the upper secondary ignition point ($I_2$) was rounded apparently. However, there were cases that the first ignition turning point was not observed clearly when the combustion products were mixed in the air stream as shown in Fig. 2. Thus, the secondary ignition point was selected as an ignition point in this study. The ignition temperature was defined by the temperature of air stream at the nozzle exit boundary. The difference in the global strain rates at the two ignition turning points, $I_1$ and $I_2$, was not so large because the horizontal axis represents the log scale value of the global strain rate.

Fig. 3 shows the ignition temperature of CH$_4$ versus the global strain rate. The ignition temperature increases with the global strain rate for a fixed dilution rate. Furthermore, the ignition temperature increases with the dilution rate for a fixed global strain rate. For large global strain rates, the increase rate of the ignition temperature further increases compared to the increase rate of dilution rate. As expected, these results mean that the ignition temperature increases as the dilution rate of combustion products becomes larger. It can be identified that the ignition of CH$_4$ is much more difficult as both the global strain rate and the dilution rate increase. For clarity, those ignition temperatures and trend obtained in this study will be validated using the experimental results in near future.
Figure 1. Response of the maximum H mole fraction to the global strain rate for pure air stream condition.

Figure 2. Response of the maximum H mole fraction to the global strain rate for case that the combustion products are mixed in the air stream at $T_0=1100$ K.

Figs 4 through 8 show the flame structure and pollutant emission characteristics of CH$_4$ nonpremixed flame ignited by hot air mixed with combustion products. Fig. 4 shows the distributions of the temperature and axial velocity with the variation of the dilution rate at $a_g = 1000$ (1/s). The air stream temperature was determined to be over 10 K than the ignition temperature for given global strain rate and dilution rate shown in Fig. 3. The volumetric percentage of oxygen in the air stream was 21% for pure air and 10.5% at $\Omega = 0.5$, respectively (Table 1). The peak flame temperature decreases and the difference between the
peak and supplying air stream temperatures decreases as the dilution rate becomes larger. The peak flame temperature of $\Omega = 0.5$ was about 2000 K. Those peak flame temperatures and differences between the peak and supplying air stream temperatures for various dilution rate are still higher compared to that of typical high-temperature air combustion. The results of Fig. 4 imply that proper high-temperature air combustion in F/BA region cannot be observed at $\Omega \leq 0.5$.

Figs. 5 and 6 show the distributions of NO and CO mole fraction for the same condition to that of Fig. 4. The concentrations of those two species were much reduced with increasing the dilution rate. Especially NO for pure air was remarkably reduced to $1/8$ compared to the case of $\Omega = 0.5$. Simultaneous increment in the supplying temperature of air stream and the dilution rate of combustion products are still effective for the NO and CO reduction in F/BA region. However, as shown in Fig. 4, the peak temperature and differences between the peak and supplying air stream temperatures are still high, thus, we needs to increase the dilution rate of combustion products in the air stream for the further reduction of the pollutants.

Table 1. Composition of the main species in the air stream with the variation of dilution rate.

<table>
<thead>
<tr>
<th>species</th>
<th>$\Omega=0.1$</th>
<th>$\Omega=0.2$</th>
<th>$\Omega=0.3$</th>
<th>$\Omega=0.4$</th>
<th>$\Omega=0.5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>O2</td>
<td>0.189</td>
<td>0.168</td>
<td>0.147</td>
<td>0.126</td>
<td>0.105</td>
</tr>
<tr>
<td>N2</td>
<td>0.782</td>
<td>0.774</td>
<td>0.766</td>
<td>0.758</td>
<td>0.750</td>
</tr>
<tr>
<td>CO</td>
<td>0.001</td>
<td>0.002</td>
<td>0.003</td>
<td>0.004</td>
<td>0.005</td>
</tr>
<tr>
<td>CO2</td>
<td>0.009</td>
<td>0.018</td>
<td>0.027</td>
<td>0.036</td>
<td>0.045</td>
</tr>
<tr>
<td>H2O</td>
<td>0.019</td>
<td>0.038</td>
<td>0.057</td>
<td>0.076</td>
<td>0.095</td>
</tr>
</tbody>
</table>

Figure 3. Ignition temperature of CH₄ versus the global strain rate.
In order to compare the pollutant emission quantitatively, emission indices for NO, NO\textsubscript{2}, CO and CO\textsubscript{2} were presented in Figs. 7 and 8. EINO decreased remarkably but EINO\textsubscript{2} increased gradually with increasing the dilution rate for the fixed global strain rate. Consequently, total EINO\textsubscript{x} (EINO + EINO\textsubscript{2}) decreased with increasing the dilution rate since EINO\textsubscript{2} was very small compared to the EINO. It should be noted that, however, the EINO at $\Omega \leq 0.5$ in F/BA region was still higher than that of usual low-temperature air nonpremixed flame [10] or BF/A region [3]. In Fig. 8, EICO decreased slightly and EICO\textsubscript{2} increased gradually with increasing the dilution rate. This result implies that the BF/A region of high-
temperature air combustor is still effective in reducing the CO emission by increasing the dilution rate but is not effective in reducing the CO₂ emission even though the dilution rate increases.

![Figure 6. Distribution of CO mole fraction CH₄ nonpremixed flame ignited by hot air with the variation or dilution rate at aₚ=1000 (1/s).](image)

![Figure 7. Variation of emission indices for NO and NO₂ with dilution rate at aₚ=1000 (1/s).](image)
Conclusions
Ignition of CH$_4$ by counterflowing hot air diluted with combustion products was investigated numerically to examine the combustion characteristics in the region between fresh fuel and diluted air with burnt gases of high-temperature air combustor. Flame structure and emission characteristics of CH$_4$ nonpremixed flame ignited by the diluted hot air were also investigated. The main results are as follows:

Ignition temperatures of CH$_4$ by hot air in the function of the global strain rate were obtained using a flame-controlling continuation method. The ignition temperature increases with the global strain rate for a fixed dilution rate. The ignition temperature also increased with the dilution rate for a fixed global strain rate. For large global strain rates, the increase rate of the ignition temperature further increased compared to the increase rate of dilution rate.

EINOx in F/BA region decreased remarkably with increasing the dilution rate for the fixed global strain rate. The EINOx for the condition considered in this study ($\Omega \leq 0.5$) was still higher than that of usual low-temperature air nonpremixed combustion or BF/A region. Furthermore, EICO decreased slightly but EICO$_2$ increased gradually with increasing the dilution rate. It was identified that the BF/A region of high-temperature air combustor contributes to the reduction of CO emission but is not effective in reducing the CO$_2$ emission even by increasing the dilution rate.

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Nomenclature

\begin{align*}
d_g & \quad \text{Global strain rate} \\
\Omega & \quad \text{Dilution rate}
\end{align*}
References


