Ignition enhancement of ethylene/air by NO\textsubscript{x} addition

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Abstract
Recently, non-equilibrium plasma assisted combustion (PAC) has been found to be promising in reducing the ignition delay time in hypersonic propulsion system. NO\textsubscript{x} produced by non-equilibrium plasma can react with intermediates during the fuel oxidation process and thereby has influence on the combustion process. In this study, the effects of NO\textsubscript{x} addition on the ignition process of both the homogeneous ethylene/air mixtures and the non-premixed diffusion layer are examined numerically. The detailed chemistry for ethylene oxidization together with the NO\textsubscript{x} sub-mechanism is included in the simulation. Reaction path analysis and sensitivity analysis are conducted to give a mechanistic interpretation for the ignition enhancement by NO\textsubscript{x} addition. It is found that for both the homogenous and non-premixed ignition processes at normal and elevated pressures, NO\textsubscript{2} addition has little influence on the ignition delay time while NO addition can significantly promote the ignition process. The ignition enhancement is found to be caused by the promotion in hydroxyl radical production which quickly oxidizes ethylene. The promotion in hydroxyl radical production by NO addition is achieved in two ways: one is the direct production of OH through the reaction HO\textsubscript{2} + NO = NO\textsubscript{2} + OH, and the other is the indirect production of OH through the reactions NO + O\textsubscript{2} = NO\textsubscript{2} + O and C\textsubscript{2}H\textsubscript{4} + O = C\textsubscript{2}H\textsubscript{3} + OH. Moreover, it is found that similar to the homogeneous ignition process, the acceleration of the diffusion layer ignition is also controlled by the reaction HO\textsubscript{2} + NO = NO\textsubscript{2} + OH.

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1. Introduction
In scramjet engines for hypersonic aircraft, the combustion resistance time is usually within a few min-seconds. In order to achieve efficient propulsion, fuel vaporization, mixing between fuel and air, ignition, and complete combustion should happen within such a short period. Therefore, ignition and combustion enhancement is of critical importance for the development of hypersonic propulsion system. Recently, non-equilibrium plasma assisted combustion (PAC) has been found to be promising in reducing the ignition delay time and enhancing the flame stabilization and propagation in hypersonic propulsion system.\textsuperscript{1–3} The ignition delay time can be greatly reduced with the help of non-equilibrium plasma by increasing both the local temperature and the radical concentration.\textsuperscript{4,5} In the literature, there have been extensive studies on PAC. For examples, Ombrello et al. found that NO\textsubscript{x} generated by non-equilibrium plasma can significantly reduce the ignition delay time of methane.\textsuperscript{6,7} Takita et al. numerically investigated the influence of adding radicals...
(H, O, OH) on the ignition process of hydrogen and methane, and the results showed that adding radicals can accelerate the chain initial reactions as well as the chain branching reactions, thus reducing the ignition delay time. However, most of the previous studies only demonstrated the performance of non-equilibrium plasma in combustion enhancement. In order to give a mechanistic interpretation for the underlying mechanisms of PAC, further research based on simulations including detailed chemistry and reaction path analysis is required.

The objectives of this study are to assess the effects of NO addition (which can be produced by non-equilibrium plasma) on the ignition of the ethylene/air mixtures and to interpret the chemical kinetics involved in the ignition enhancement by NO addition. Ethylene is chosen as the fuel because it is the main product of the thermal dissociation of the large hydrocarbon fuels which are widely used as the propulsion energy source. In the following text, the numerical models and specifications are presented first. Then, the effects of NO addition on the homogeneous and the non-premixed diffusion layer ignition processes are assessed, and the chemical kinetics involved in the ignition enhancement is discussed. Finally, the main conclusions are summarized.

2. Numerical methods and specifications

In order to assess the kinetic effects of NO addition on the ignition of the premixed ethylene/air mixture, the homogeneous ignition process at constant pressure is simulated. The governing equations for the temperature T and the mass fraction of the kth species \( Y_k \) are

\[
\frac{dT}{dt} = -\sum_{k=1}^{n} \left( h_k \omega_k \frac{W_k}{\rho} \right)
\]

\[
\frac{dY_k}{dt} = \frac{\omega_k W_k}{\rho} \quad \text{for } k = 1, 2, \ldots, n
\]

where \( t, \rho, c_p, h_k, \omega_k, W_k \) and \( n \) denote, respectively, the time, density, specific heat at constant pressure, enthalpy, production rate and molecular weight of \( k \)th species, and the total number of species. The chemical reaction rates and the thermal properties under different conditions (temperature, pressure, and composition) are calculated using the CHEMKIN package. The ordinary differential Eqs. (1) and (2) are integrated implicitly using the VODE solver.

The homogeneous ignition process is purely controlled by chemical kinetics in which there are no mass and heat transport. However, in practical PAC process, the ignition is also affected by mixing of fuel and air as well as heat conduction. In order to assess the ignition enhancement in circumstances where transport processes happen, we study the non-premixed ignition process in a diffusion layer configuration. The in-house code A-SURF is employed to simulate the non-premixed ignition process in the diffusion layer between the cold fuel and hot air. A-SURF solves the conservation equations of one-dimensional, compressible, multi-component, reactive flow using the finite volume method. The CHEMKIN package is incorporated into A-SURF to calculate the temperature- and component-dependent thermodynamic and transport properties as well as the chemical reaction rates. A-SURF has been successfully used in our previous studies.

3. Results and discussion

3.1. Effects of NO addition on homogeneous ignition process

There are many different ways to define the ignition delay time, \( \tau \), in the literature. In this study, the definition of ignition delay time is based on the evolution of the temperature as a function of time, \( T(t) \), and \( \tau \) is the time at which the peak value of the rate in the change of the temperature (\( dT/dt \)) occurs (which corresponds to the maximum heat release rate). It is also acceptable to define the ignition delay time as the time when the temperature reaches certain degree above the initial temperature \( T_0 \), e.g., \( T_0 + \Delta T \), or as the sudden rise in the OH concentration, where OH represents the excited electronic state of OH radical as commonly used in shock-tube experiments. As demonstrated in our previous study, the ignition delay times determined in these three ways are very close. Therefore, different definitions of the ignition delay times can be used interchangeably since they are quantitatively similar.

Fig. 1 demonstrates the effects of NO addition on the ignition delay time of the stoichiometric ethylene/air mixtures at different initial temperatures and pressures \( p \). It is seen that at both atmospheric and elevated pressures (10^5 and 10^6 Pa), NO addition in the amount of 0.5% vol. can reduce the ignition delay time by one order of magnitude when the initial temperature is in the range of 1000–1200 K. Compared to NO, NO2 addition is shown to have much weaker influence on the ignition delay time. Therefore, NO is much more effective than NO2 in terms of ignition enhancement for ethylene/
air mixtures. Moreover, Fig. 1 indicates that the ignition enhancement by NO addition decreases with the initial temperature. It is seen that when the initial temperature is above 1400 K, the ignition happens within 10^{-4} s and NO addition has little influence on the ignition delay time.

There are different reaction mechanisms for NOx available in the literature. To ensure that our conclusions do not depend on the NOx sub-mechanism, the calculations are repeated and compared based on the USC-Mech II mechanism integrating with two other NOx sub-mechanisms from, respectively, the SanDiego-Mech 2005 and the Konnov’s mechanism. As shown in Fig. 1, the results from these different mechanisms agree quite well with each other. This demonstrates the validity of the kinetic model used in the present work. Therefore, in the following only the NOx sub-mechanism obtained from the GRI-Mech 3.0 is used in simulation.

Fig. 2 shows the change of the ignition delay time with the amount of NOx addition. Similar to the results in Fig. 1, NO2 addition is shown to have little influence on the ignition delay time, while NO addition is able to significantly promote the ignition process and to reduce the ignition delay time by more than one order of magnitude. Moreover, unlike the initial temperature which greatly affects the ignition enhancement at a given amount of NO addition (as shown in Fig. 1), the pressure is shown in Fig. 2 to have little influence: the reduction in the ignition delay time at a given amount of NO addition at normal pressure (10^5 Pa) is nearly the same as that at elevated pressure (10^6 Pa).

It is observed in Fig. 2 that too much NOx addition can prohibit the ignition process resulting in longer ignition delay time, especially for NO2 addition. In practical PAC process, the NOx concentration is usually within several thousand ppm. Therefore, according to Fig. 2, the ignition process should be always enhanced by NOx produced by non-equilibrium plasma.

3.2. Effects of NOx addition on non-premixed ignition process

The non-premixed ignition occurs in a diffusion layer between the static cold fuel (T = 300 K) and hot air (T = 1100 K). Fig. 3(a) shows the evolution of the temperature distribution during the non-premixed ignition process without any NOx addition. The initial (t = 0 ms) temperature and the composition concentration distributions are in the form of a step-function with the discontinuity located at x = 10.0 cm. After the contact of the hot air with the cold fuel, heat conduction and mass diffusion occur and a mixing layer is developed. Meanwhile, chemical reactions take place during the mixing process. Since the reaction rate is more sensitive to the temperature than to the concentrations of the reactants, the ignition kernel is first found in the hot air side, as shown in Fig. 3(a). Moreover, it is observed in Fig. 3(a) that the ignition kernel propagates towards the cold fuel side. This is due to the facts that the small amount of fuel diffused into the hot air can be consumed in the ignition kernel, and that there is more fuel diffused into the air side at the position closer to the initial contact surface (x = 10.0 cm).

The evolution of the maximum temperature during the non-premixed ignition process with different amounts of NOx addition is plotted in Fig. 3(b). It is seen that the non-premixed ignition process is also strongly affected by NO addition. Similar to the homogeneous ignition process, the non-premixed
ignition delay time is defined as the time when the peak value of the rate in the change of the maximum temperature \((dT_{\text{max}}/dt)\) occurs.

Fig. 4 demonstrates the effects of NO\(_x\) addition on the non-premixed ignition delay time in a diffusion layer configuration. Similar to the results in Fig. 2 for the homogeneous ignition process, Fig. 4 indicates that NO addition can greatly promote the non-premixed ignition process and reduce the ignition delay time by more than one order of magnitude. Unlike NO, adding NO\(_2\) is shown to have little influence on the non-premixed ignition delay time. It is noted that the ignition delay time of the non-premixed ignition process is much larger (by a factor around ten) than that of the homogeneous ignition process, which can be observed by the comparison between Figs. 4 and 2. This is due to the facts that the cold fuel and hot air need to mix with each other by heat and mass diffusion in the non-premixed ignition process, and that the average temperature in the non-premixed ignition process is lower than the homogeneous ignition process (which is 1100 K in Fig. 2).

In order to explain the change of the ignition delay time with NO addition observed in Figs. 1–4, reaction path analysis and sensitivity analysis are conducted in the next subsection based on the homogeneous system to understand the kinetics involved in the ignition enhancement.

3.3. Chemical kinetics involved in ignition enhancement

Chemical reaction path analysis is conducted for the homogeneous ignition process of stoichiometric ethylene/air mixture at \(T_o = 1100\) K and \(p = 10^5\) Pa without and with 0.1% vol. NO addition. Fig. 5 shows the evolution of the molar fraction of major radicals (OH, O, HO\(_2\), and C\(_2\)H\(_3\)) as well as NO and NO\(_2\) during the homogeneous ignition process. The evolution of the molar fraction of H radical is close to that of O radical and thereby is not shown in Fig. 5 for clear demonstration. It is seen that with 0.1% vol. NO addition, the radical pool is much more easily generated and thereby the ignition process is greatly accelerated. Without NO addition, the OH and O radicals can be built up only after \(t > 10^{-3}\) s. However, with 0.1% vol. NO addition, the OH and O radicals can be generated much earlier which results in the reduction of the ignition delay time. Moreover, it is observed in Fig. 5(b) that NO is converted into NO\(_2\) during an initial period. As will be explained later, this process in fact plays an essential role in promoting the ignition process.

It is well-known that in the homogeneous ignition process of the ethylene/air mixture, the ethylene is first oxidized to
ethenyl (C₂H₃) through the reaction \( \text{C}_2\text{H}_4 + \text{O}_2 = \text{C}_2\text{H}_3 + \text{HO}_2 \). The rates of this reaction and the \( \text{C}_2\text{H}_3 \) oxidation reactions basically control the ignition process.⁵ Therefore, we first investigate the generation path of \( \text{C}_2\text{H}_3 \) with and without NO addition. We introduce the following definition of the contribution of the \( \text{i} \)th reaction to the concentration change of a certain intermediate species:²⁹,³⁰

\[
C_i = \frac{f_i}{\sum_{i=1}^{m} |f_i|}
\]

where \( f_i \) represents the rate of change in the concentration of the considered species due to the \( \text{i} \)th reaction, and \( m \) is the total number of elementary reactions. Therefore, the positive (negative) value of \( C_i \) indicates that the considered species is generated (consumed) by the \( \text{i} \)th reaction. It is noted that the values of \( C_i \) at different time cannot be directly compared since the reaction rates (and so do the numerator and enumerator in Eq. (3)) change greatly with time in the ignition process. Only the comparison between values of \( C_i \) for different reactions at the same time is meaningful.

The major reactions related to the production and consumption of \( \text{C}_2\text{H}_3 \) during the ignition process of atmospheric ethylene/air mixtures at initial temperature \( T_u = 1000 \text{ K} \) are plotted in Fig. 6. It is seen that without NO addition, \( \text{C}_2\text{H}_3 \) is mainly produced by the reaction between \( \text{C}_2\text{H}_4 \) and \( \text{O}_2 \) at \( t < 6 \times 10^{-5} \text{ s} \), after \( \text{C}_2\text{H}_4 \) is mainly oxidized by hydroxyl radical \( \text{OH} \) through the reaction \( \text{C}_2\text{H}_4 + \text{OH} = \text{C}_2\text{H}_3 + \text{H}_2\text{O} \). However, with 0.1\% vol. NO addition, the reaction between \( \text{C}_2\text{H}_4 \) and \( \text{OH} \) is greatly advanced and becomes dominant in the oxidation of \( \text{C}_2\text{H}_4 \) when \( t > 4 \times 10^{-6} \text{ s} \). Therefore, the ignition enhancement by NO addition is mainly due to the earlier appearance of \( \text{OH} \) radical which greatly accelerates the oxidation of \( \text{C}_2\text{H}_4 \). Moreover, the comparison between Fig. 6(a) and (b) indicates that NO addition has little influence on the further oxidation path of \( \text{C}_2\text{H}_3 \).

Since the appearance of \( \text{OH} \) radical can greatly accelerate the oxidation of \( \text{C}_2\text{H}_4 \), we investigate the reaction path of \( \text{OH} \) radical and the results are presented in Fig. 7. It is seen that without NO addition, \( \text{OH} \) is mainly produced through the reaction \( \text{C}_2\text{H}_4 + \text{O} = \text{C}_2\text{H}_3 + \text{OH} \) at a very small reaction rate due to the extremely low concentration of \( \text{O} \) radical (see Fig. 5). However, with 0.1\% vol. NO addition, as shown in Fig. 7(b), the \( \text{OH} \) radical can be generated through the reaction \( \text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH} \) at a much faster reaction rate since the concentration of \( \text{HO}_2 \) is several orders higher than that of \( \text{O} \) radical (see Fig. 5). Therefore, NO addition can greatly enhance the production of \( \text{OH} \) radical via the reaction \( \text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH} \), in which \( \text{HO}_2 \) is mainly produced by the reaction \( \text{C}_2\text{H}_4 + \text{O}_2 = \text{C}_2\text{H}_3 + \text{HO}_2 \) as demonstrated in Fig. 6. Similar analysis have been done to study contributions to \( \text{OH} \) accumulation at the same conditions, and the results, as shown in Fig. 7, demonstrated that the main consumption path of \( \text{OH} \) radical is to oxidize \( \text{C}_2\text{H}_4 \) through the reaction \( \text{C}_2\text{H}_4 + \text{OH} = \text{C}_2\text{H}_3 + \text{H}_2\text{O} \).

Fig. 8 demonstrates the main generation and consumption reactions of oxygen radical \( \text{O} \) during the ignition process of atmospheric ethylene/air mixtures at initial temperature \( T_u = 1000 \text{ K} \) with and without NO addition. When NO is not added, Fig. 8(a) indicates that \( \text{O} \) radical is mainly produced by the reaction \( \text{C}_2\text{H}_3 + \text{O}_2 = \text{CH}_2\text{CHO} + \text{O} \). When
0.1% vol. NO is added, Fig. 8(b) shows that before \( t = 10^{-7} \) s, O radical is mainly produced by the reaction \( \text{NO} + \text{O}_2 = \text{NO}_2 + \text{O} \), which is much faster than the reaction \( \text{C}_2\text{H}_3 + \text{O}_2 = \text{CH}_2\text{CHO} + \text{O} \) since the concentration of NO is much higher than that of \( \text{C}_2\text{H}_3 \) (see Fig. 5). Therefore, with the appearance of NO, O radical can be quickly produced through the reaction \( \text{NO} + \text{O}_2 = \text{NO}_2 + \text{O} \), after which O radical reacts with \( \text{C}_2\text{H}_4 \) to produce OH through the reaction \( \text{C}_2\text{H}_4 + \text{O} = \text{C}_2\text{H}_3 + \text{HO} \).

The reaction rates of ethenyl involved reactions during the ignition process of atmospheric ethylene/air mixtures at initial temperature \( T_u = 1000 \) K are plotted in Fig. 9. The solid lines denote results with 0.1% vol. NO addition while the dashed lines without NO addition. The reaction rate of \( \text{C}_2\text{H}_4 + \text{O}_2 = \text{C}_2\text{H}_3 + \text{HO}_2 \) is expanded ten times and this reaction is shown to be the chain initiation reaction in the ignition process without NO addition. When NO is added, a great amount of OH generated through the reaction \( \text{NO} + \text{HO}_2 = \text{OH} + \text{NO}_2 \) can participate in the fast reaction \( \text{C}_2\text{H}_4 + \text{OH} = \text{C}_2\text{H}_3 + \text{H}_2\text{O} \), and thereby accelerates the oxidation of ethylene molecule. The O radical is shown to oxidize \( \text{C}_2\text{H}_4 \) through the reactions \( \text{C}_2\text{H}_4 + \text{O} = \text{C}_2\text{H}_3 + \text{OH} \) and \( \text{C}_2\text{H}_4 + \text{O} = \text{CH}_4 + \text{HCO} \).

Therefore, according to the results shown in Figs. 6-9, the ignition enhancement by NO addition is caused by the promotion in OH radical production which quickly oxidizes \( \text{C}_2\text{H}_4 \) through the reaction \( \text{C}_2\text{H}_4 + \text{OH} = \text{C}_2\text{H}_3 + \text{H}_2\text{O} \) (see Fig. 6). The promotion in OH production by NO addition is achieved in two ways: one is the direct production of OH through the reaction \( \text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH} \) (see Fig. 7), and the other is the indirect production of OH through the reactions \( \text{NO} + \text{O}_2 = \text{NO}_2 + \text{O} \) and \( \text{C}_2\text{H}_4 + \text{O} = \text{C}_2\text{H}_3 + \text{OH} \) (see Fig. 8).

In order to further reveal the elemental reactions involved in the ignition enhancement by NO addition, sensitivity analysis is conducted for the homogeneous ignition process of atmospheric ethylene/air mixtures at initial temperature \( T_u = 1000 \) K with and without 0.1% vol. NO addition. The logarithmic sensitivities of different reactions are compared in Fig. 10.

It is observed that without NO addition, the ignition delay time is mainly affected by the reactions involved in the oxidation of \( \text{C}_2\text{H}_4 \) and \( \text{C}_2\text{H}_3 \) by \( \text{HO}_2 \) and \( \text{O}_2 \), respectively. However, in the case of 0.1% vol. NO addition, the sensitivity of these reactions is reduced indicating that the ignition process is affected by other elementary reactions. This is consistent with the results shown in Figs. 6-9. Moreover, Fig. 10 indicates that the elemental reactions \( \text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH} \) and \( \text{C}_2\text{H}_4 + \text{OH} = \text{C}_2\text{H}_3 + \text{H}_2\text{O} \) strongly influence the homogeneous ignition process when 0.1% vol. NO is added into the mixture. This further validates the conclusion that the ignition enhancement by NO addition is caused by the promotion in OH radical production (through reaction \( \text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH} \)) which quickly oxidizes \( \text{C}_2\text{H}_4 \) (through reaction \( \text{C}_2\text{H}_4 + \text{OH} = \text{C}_2\text{H}_3 + \text{H}_2\text{O} \)).

Fig. 11 shows the change of the normalized ignition delay time with NO addition for the homogeneous and diffusion

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**Fig. 8** Major reactions related to the production \((C_i > 0)\) and consumption \((C_i < 0)\) of O in the homogeneous ignition process of stoichiometric ethylene/air mixture.

**Fig. 9** Reaction rates of ethenyl involved elemental reactions in homogeneous ignition process of stoichiometric ethylene/air mixture without and with 0.1% vol. NO addition.

**Fig. 10** Logarithmic sensitivity analysis on ignition delay time of stoichiometric ethylene/air mixture without and with 0.1% vol. NO addition.
layer systems. The normalized ignition delay time is defined as $\tau(c)/\tau(c = 0)$, the inverse of which is the speed-up in ignition caused by NO addition. It is seen that the change of the normalized ignition delay time with the volume fraction of NO is similar for both systems. Therefore, the non-premixed ignition process is also enhanced by NO-related elementary reactions. Furthermore, Fig. 11 demonstrates the effects of the most important enhancement reaction $\text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH}$ on the normalized ignition delay time of the homogeneous and diffusion layer systems. When reaction $\text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH}$ is frozen, the normalized ignition delay times of both the homogeneous and the diffusion layer systems are close to 1.0, which indicates that the ignition cannot be promoted by NO addition. Therefore, similar to the homogeneous ignition process, the acceleration of the diffusion layer ignition is also controlled by the reaction $\text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH}$.

4. Conclusions

1. It is found that NO$_2$ addition has little influence on the ignition delay time, while NO addition can significantly promote the ignition process.

2. For the homogeneous ignition process, it is shown that the ignition enhancement by NO addition decreases with the initial temperature and that NO addition has little influence on the ignition delay time when the initial temperature is above 1400 K. For the non-premixed ignition process, the ignition delay time is much longer than that of the homogeneous ignition process. Nevertheless, for both the homogeneous and non-premixed ignition processes, the ignition delay time can be reduced by more than one order of magnitude when a certain amount of NO is added.

3. Reaction path analysis and sensitivity analysis are conducted for the homogeneous ignition process without and with 0.1% vol. NO addition and it is found that the ignition enhancement by NO addition is caused by the promotion in OH radical production which quickly oxidizes $\text{C}_2\text{H}_4$ through the reaction $\text{C}_2\text{H}_4 + \text{OH} = \text{C}_2\text{H}_3 + \text{H}_2\text{O}$. The promotion in OH production by NO addition is achieved in two ways: one is the direct production of OH through the reaction $\text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH}$, and the other is the indirect production of OH through the reactions $\text{NO} + \text{O}_2 = \text{NO}_2 + \text{O}$ and $\text{C}_2\text{H}_4 + \text{O} = \text{C}_2\text{H}_3 + \text{OH}$. Furthermore, we demonstrate that similar to the homogeneous ignition process, the acceleration of the diffusion layer ignition is also controlled by the reaction $\text{HO}_2 + \text{NO} = \text{NO}_2 + \text{OH}$.

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References


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