

Ignition And Flame Propagation Enhancement by Plasma Excited Oxygen: Role of Ozone

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Abstract. In this study, the effects of plasma assisted combustion on both the ignition and flame propagation processes have been investigated. Specifically, the effects of ozone have been investigated. By numerical simulations of the ignition process of plasma assisted combustion with detailed fuel and ozone kinetics, the plasma generated ozone has been found to reduce the ignition delay times. Moreover, the enhanced reactivity by ozone additions is mainly due to the chemical kinetic effect, while thermodynamics only has a secondary influence. By simulating the flame propagation of ozone additions to both hydrogen/oxygen and methane/oxygen flames, the flame propagation speeds have been found to also increase with additions of plasma generated ozone. However, the enhancement of flame propagation is less sensitive to ozone additions compared with the ignition process. The current investigation provides useful insights into plasma assisted combustion as well as guidance to new strategies for enhancing various propulsion systems.

Keywords: Plasma, Ozone, Ignition, Flame propagation.

1. Introduction

Achieving highly efficient and powerful propulsion is essential for various applications, such as jet engines, detonation engines and rockets. For the unburned hydrocarbons and emissions such as NO_x , the addition of plasma could also reduce the unburned hydrocarbons and NO_x . In the meantime, new methods for controlling the combustion processes in these advanced propulsion systems are needed to maintain steady operations. These challenges require innovative ways for enhancing combustion processes in real applications. Among various combustion enhancement and controlling methods, the *plasma assisted combustion* [1] is a promising way to improve combustion efficiency as well as provide controlling strategies for propulsion systems. For many of the high-speed propulsion systems, the flow time scale is much shorter compared with the chemical time scale, as such the high-speed flow of combustible gas cannot be fully reacted to achieve high combustion efficiencies. Active species generated in plasma could substantially reduce the chemical time scale of the system, as such it can be comparable to the flow time scale to make the combustion process more complete.

Plasma is one of four fundamental states of matter with the other three as gas, liquid, and solid. It is characterized by the presence of large portions of charged particles [2]. This makes the plasma state very reactive with all the active species such as ions and electrons. Plasma state can be achieved by heating up a neutral gas to very high temperatures, or it can be generated by using very strong electromagnetic fields. Plasma assisted combustion is the method of enhancing and controlling combustion by adding plasma into combustible mixtures. Among all the active species generated by plasma, ozone (O_3) can be generated from plasma excited oxygen. It is one of the most important species for plasma assisted combustion.

To explain how plasma addition could enhance the combustion processes, such as ignition, flame propagation, flame stabilization and so on, we introduce the Arrhenius expression [3] of chemical reaction:

$$k(T) = A \cdot \exp(-E_a/RT) \quad (1)$$

where $k(T)$ is the chemical reaction rate coefficient indicating how fast chemical reactions can happen, A is the pre-exponential factor, E_a is the activation energy, R is the universal gas constant,

and T is the temperature. Adding plasma leads to two different effects that could increase $k(T)$. The first effect is thermal *effect*, which means plasma assisted combustion could increase the heat release of the system and therefore increase the temperature T of the system. Consequently, the chemical reaction rate is faster than the situation without plasma, because the $\exp(-E_a/RT)$ term is increased. The other effect is the *kinetic effect*. This is the effect that, by the addition of active species such as free radicals, ions and electrons, new kinetic pathways can be activated, and existing pathways can be enhanced. In the Arrhenius expression, the pre-exponential factor A is then increased, and the activation energy E_a is decreased, both of which increase the reaction rate coefficient $k(T)$. However, the chemical kinetics of plasma assisted combustion is a highly complicated and coupled process. It could include multiple different pathways such as the fuel decomposition into smaller fragments, the interaction between fuel molecule and the charged particles, and so on. Specifically, the different ways that plasma can enhance combustion process is summarized in Figure 1.

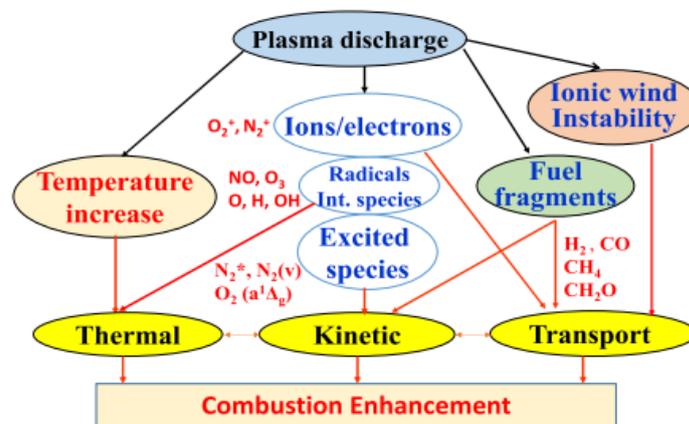


Figure 1. Different effects of plasma assisted combustion, from Ref. [1].

In literature, there have been extensive investigations on plasma assisted combustion. Ombrello *et al.* [4, 5] experimentally investigated plasma assisted flames with the emphasis on the role of plasma excitation of oxygen. They explained the effects of plasma assisted combustion based on the radicals, excited species, ions, and electrons as well as elevated temperatures due to plasma additions. Leonov and Yarantsev [6] investigated plasma induced ignition and plasma assisted combustion in high-speed flows. Recently, the enhanced flammability of swirling ammonia/air combustion using gliding arc plasma has been examined and discussed in [7]. The ignition and combustion enhancement in a cavity-based supersonic combustor by the multi-channel gliding arc plasma have been investigated in [8]. Ref. [9] numerically investigated the plasma assisted combustion in the sequential combustor. In Ref. [10], an incremental methodology has been used to build the plasma mechanism, which focuses on three aspects of plasma additions: fast gas heating, slow gas heating and radical production.

These studies mostly focus on the global responses of plasma assisted combustion. However, the detailed kinetics of plasma assisted combustion still needs more explanations. The specific role of ozone addition generated by plasma excitation has been analyzed in Ref. [11]. This study mostly focused on the plasma assisted flame processes, but the plasma assisted ignition process is not fully discussed. Moreover, these investigations also pointed out that plasma assisted combustion is a very complicated process, which couples many different factors. As such, it is difficult to explore the detailed kinetic pathways and quantify the specific effects of different influencing parameters. More systematic investigations of plasma assisted combustion, especially the key kinetic pathways, are still needed to explore the complicated couplings between plasma generated active species and combustion species.

In the current investigation, the objective is to use detailed kinetic simulations to study the underlying chemical thermodynamic and chemical kinetic effects of plasma assisted combustion.

Specifically, we will examine the role of plasma excited species on both the ignition and flame propagation phenomena for fuels typically used in propulsion such as hydrogen and methane. Ozone is one of the major species generated by plasma excitation of oxygen, so the current investigation will focus on the role of ozone as the major species for the enhancement of combustion by plasma. Using both the ignition and flame propagation simulations, the effects of plasma additions on different combustion modes could also be identified. The goal of the current investigation is to explain the phenomena of plasma assisted combustion, and as such provide new insights and guidance for the various applications of plasma assisted combustion.

2. Methods

The current study uses numerical simulations to investigate the detailed reaction system of plasma assisted combustion. We adopted the Python package Cantera [12] to perform simulations of both ignition and flame propagation. Cantera incorporates the detailed thermodynamic information of different species as well as the chemical kinetic information of both the conventional fuel oxidation and the plasma activated kinetic pathways. Furthermore, it could solve the governing equations of mass conservation, species conservation and energy conservation with different numerical methods. From the computations of Cantera, global combustion responses such as the ignition delay time and the laminar flame speed can be obtained for typical fuel systems such as hydrogen and methane.

For the ignition process, the computer program solves the ordinary differential equations (ODEs) controlling the time evolutions of species concentrations and temperature. Both the constant-volume and constant-pressure reactors have been used as computational models. As such the time evolutions of different species as well as the temperature can be obtained from the numerical simulations. The detailed kinetics of hydrogen and methane has been modeled by the mechanism developed in [13], which includes the species information and detailed kinetic pathways for both hydrogen and methane combustion. The specific kinetic pathways as well as thermodynamic data of plasma generated species ozone has been added into the computational model using the model in [14], which include the elementary reactions of ozone decomposition as well as ozone reactions with other species. These models have been validated against various experimental data.

For the flame propagation process, the one-dimensional flame structure has been simulated with Cantera. The governing equations of mass conservation, species conservation, and energy conservation are solved numerically by using finite different methods. In addition to the chemical model, the simulation of flame propagation also needs to include the transport model [15], which describes the diffusion process of species as well as the heat conduction process. For the oxidation of both hydrogen and methane, the kinetic model in [16] has been used. This model consists of both kinetic information and the transport model for hydrogen and methane combustion.

3. Plasma Assisted Ignition

The effects of plasma excited oxygen on the ignition of typical fuels can be analyzed in two aspects, namely the effect of chemical thermodynamics and the effect of chemical kinetics. For the ignition processes, chemical thermodynamics plays a role as it will affect the temperature of the oxidation of fuel with ozone additions. Moreover, the chemical kinetics would substantially influence the ignition delay time, which is controlled by the various elementary reactions involved. In the following two subsections, the separated roles of chemical thermodynamics and chemical kinetics will be explored using the numerical simulation results.

3.1. Effect of Chemical Thermodynamics

The effect of chemical thermodynamics will be explored in this section by the calculations of chemical equilibrium. The adiabatic flame temperature is adopted as the global response to investigate the role of ozone addition on the combustion processes of typical fuels.

Here, we adopt the concept of equivalence ratio to quantify the percentages of fuel and oxidizer in the mixture. The equivalence ratio is defined as:

$$\phi = \left(\frac{[F]}{[O]}\right) / \left(\frac{[F]}{[O]}\right)_{st} \quad (2)$$

where the $\left(\frac{[F]}{[O]}\right)$ denotes the mole concentration ratio of fuel and oxidizer, and the subscript st indicates the stoichiometric condition. Taking hydrogen and oxygen as an example, at the stoichiometric condition the reaction happens as $2H_2 + O_2 = 2H_2O$, for which the $\left(\frac{[F]}{[O]}\right)_{st} = 2$. If the oxygen is abundant, the mixture is called a lean mixture and $\phi < 1$; if the fuel is abundant, the mixture is called a rich mixture and $\phi > 1$.

First, we investigate the effect of ozone addition on the adiabatic flame temperatures of hydrogen/air mixtures. The initial condition of the calculation is that the pressure (P) is at 1 atm and the initial temperature (T_u) is at 300 K.

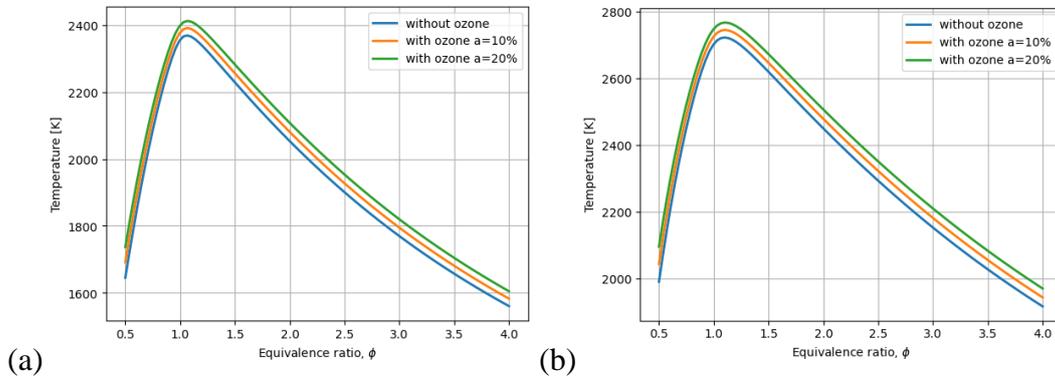


Figure 2. Adiabatic flame temperatures of hydrogen/air mixtures with different ozone concentrations at $P = 1 \text{ atm}$, $T_u = 300 \text{ K}$ for (a) constant pressure and (b) constant volume conditions.

Figure 2 demonstrates the adiabatic flame temperatures of hydrogen/air mixtures with different ozone concentrations for both constant pressure and constant volume conditions. The concentration of ozone is quantified by the amount of oxygen that has been transferred into ozone by plasma excitation. Here, the parameter a is defined as:

$$a = Y_{O_3} / (Y_{O_2} + Y_{O_3}) \quad (3)$$

where, Y_{O_3} is the ozone mass fraction after plasma excitation of oxygen and Y_{O_2} is the oxygen mass fraction. Therefore, the situation of $a = 0$ indicates that there is no plasma excited oxygen into ozone.

It can be seen that with more oxygen in the air being excited by plasma into ozone, the corresponding adiabatic flame temperature will be higher. This temperature enhancement is potentially the factor that plasma excited oxygen could enhance combustion of hydrogen. The Arrhenius law shows that the higher temperature will lead to the larger chemical reaction rate coefficient. The increased temperature by plasma excited oxygen could make the combustion process faster as the rates of chemical reactions are enhanced at elevated temperatures. Furthermore, it is observed that the constant pressure cases have lower flame temperatures compared with the constant volume cases. The reason is that part of the heat release in the constant pressure cases has been used to do the expansion work, as such its adiabatic temperature is lower than the constant volume cases.

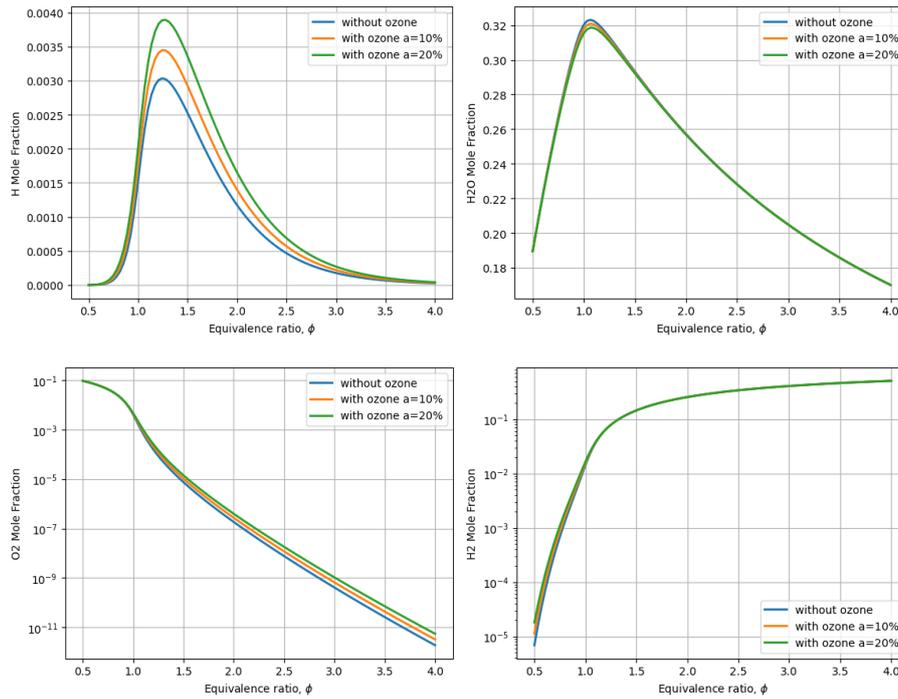


Figure 3. Equilibrium mole fractions of H radical, H₂O, O₂ and H₂ with different ozone concentrations as a function of equivalence ratio.

Figure 3 shows the variations of the equilibrium mole fractions of several key species as a function of the equivalence ratio. The results demonstrate that the H radical and H₂O profiles have the maximum values near the unity equivalence ratio, but the H radical profiles have more substantial differences for different ozone additions. This finding indicates that the equilibrium concentrations of free radicals are more likely to be modified with ozone additions, while the H₂O, as the major product, is less influenced by the ozone additions. For the fuel (hydrogen) and oxidizer (oxygen), they are less affected by the ozone additions, and their mole fraction profiles show opposite trends for the change of equivalence ratios.

Next, we investigate the effect of ozone addition on the adiabatic flame temperatures of hydrogen/oxygen and methane/oxygen mixtures. It shall be noted that for rocket propulsion normally pure oxygen is used as the oxidizer and the typical fuels used are hydrogen and methane. Especially in recently developed rockets, methane has been considered as a promising fuel for propulsion. We now compare the results of using hydrogen and methane as fuels at constant pressure conditions. Here, the initial condition of the calculation is that the pressure (P) is at 1 atm and the initial temperature (T_u) is at 300 K.

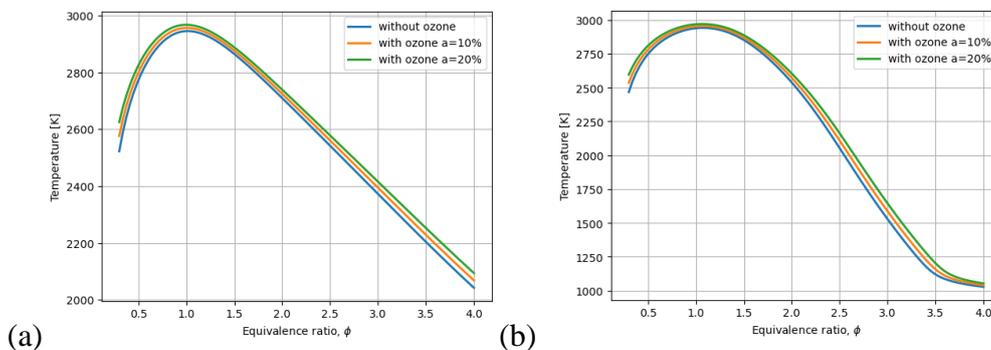


Figure 4. Adiabatic flame temperatures of (a) hydrogen/oxygen and (b) methane/oxygen mixtures with different ozone concentrations as a function of equivalence ratio.

In Figure 4, it is noted that for both the hydrogen/oxygen mixtures and the methane/oxygen mixtures, the adiabatic flame temperatures are higher than these of the hydrogen/air mixtures, which

has been shown in Figure 2. For the pure oxygen situations, the flame temperature can reach the maximum value about 3000 K. However, for the previous cases with air as the oxidizer, the maximum temperature is only around 2400 K. Such finding indicates the additional nitrogen in air could make the flame temperature lower.

Next, the comparison between the two different fuels, namely hydrogen and methane, shows that the maximum flame temperatures for both fuels are nearly the same, with the values slightly less than 3000 K. However, the difference is that methane/oxygen mixtures have a lower flame temperature on the rich side of the equivalence ratios, which is due to the formation of incomplete combustion products, such as CO. The hydrogen/oxygen mixtures have a higher temperature on the rich side compared with the methane/oxygen mixtures as it does not contain carbon and cannot decompose into CO.

As for the effect of ozone additions, the results with different amounts of ozone have relatively small differences, specifically for cases with methane as the fuel. Such observation indicates that the chemical thermodynamic effect is probably not the most important effect in terms of the enhancements by plasma additions.

3.2. Effect of Chemical Kinetics

In this subsection, we examine the effects of chemical kinetics on the plasma assisted ignition process. For the selection of fuels, we adopt hydrogen and methane as the typical fuels, which have been used in real applications, such as rocket engines. Different from the previous subsection, which discusses the chemical thermodynamic effects, we now investigate the chemical kinetic processes. For the chemical thermodynamic effects, we only concerned about the final equilibrium states of combustion, but for the chemical kinetic effects, we examine the time evolution of the reaction system, which offers more detailed information on how fast the ignition process happens. Now, we examine the time-evolution profiles the ignition process with and without the plasma excited species, as such the role of active species on the reactivity of the system can be identified.

We first examine the temperature and species profiles for the ignition process with and without the ozone addition. Hydrogen has been used as the fuel, and pure oxygen has been selected as the oxidizer. The constant-pressure ignition processes have been simulated with the equivalence ratio $\phi = 1.0$, initial temperature $T_u = 1000 K$ and the pressure $P = 1 atm$. The amount of ozone added into the mixture is $a = \frac{Y_{O_3}}{Y_{O_2} + Y_{O_3}} = 0.1\%$.

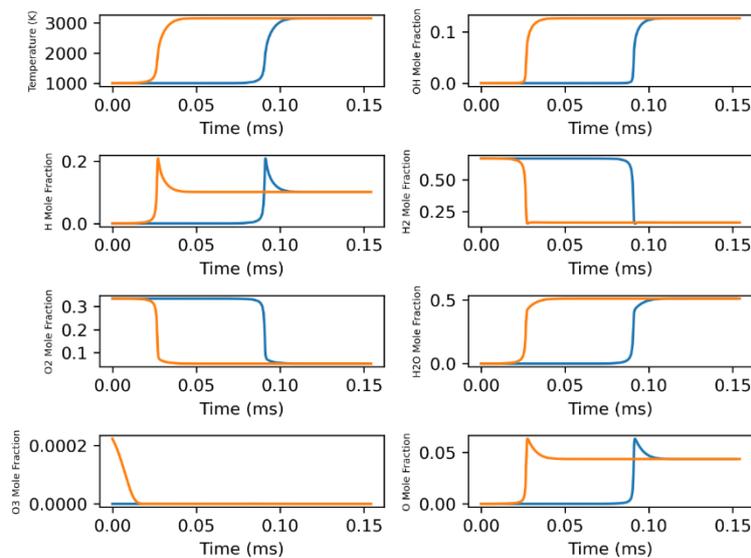


Figure 5. Temperature and species profile of hydrogen/oxygen ignition with and without ozone addition at equivalence ratio $\phi = 1.0$, $P = 1 atm$ and $T_u = 1000 K$. (Blue lines: without ozone; orange lines: with ozone)

As demonstrated in Figure 5, the blue lines show the results without ozone addition and the orange lines show the results with ozone addition. The ignition process becomes much faster for the situation with ozone addition compared with the that without ozone addition, even with only $a = 0.1\%$ of ozone addition. It is noted that there exists a moment when the temperature of the system suddenly increases, and the fuel and oxygen mole fractions suddenly decrease. Such time is defined as the ignition delay time of the ignition process. It can be quantified that after adding in ozone into the system the ignition delay time changes from 0.091 ms to 0.027 ms . Such findings suggest that small amounts of ozone addition can substantially accelerate the ignition of hydrogen.

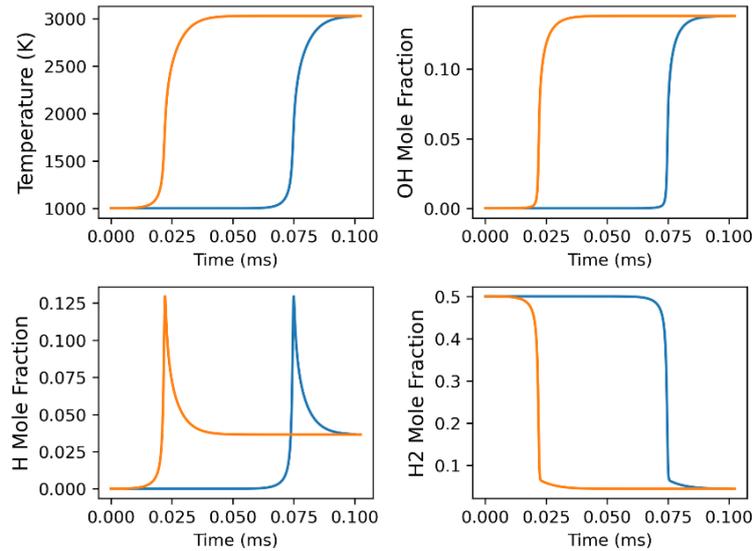


Figure 6. Temperature and species profile of hydrogen/oxygen ignition with and without ozone addition at equivalence ratio $\phi = 0.5$, $P = 1\text{ atm}$ and $T_u = 1000\text{ K}$. (Blue lines: without ozone; orange lines: with ozone)

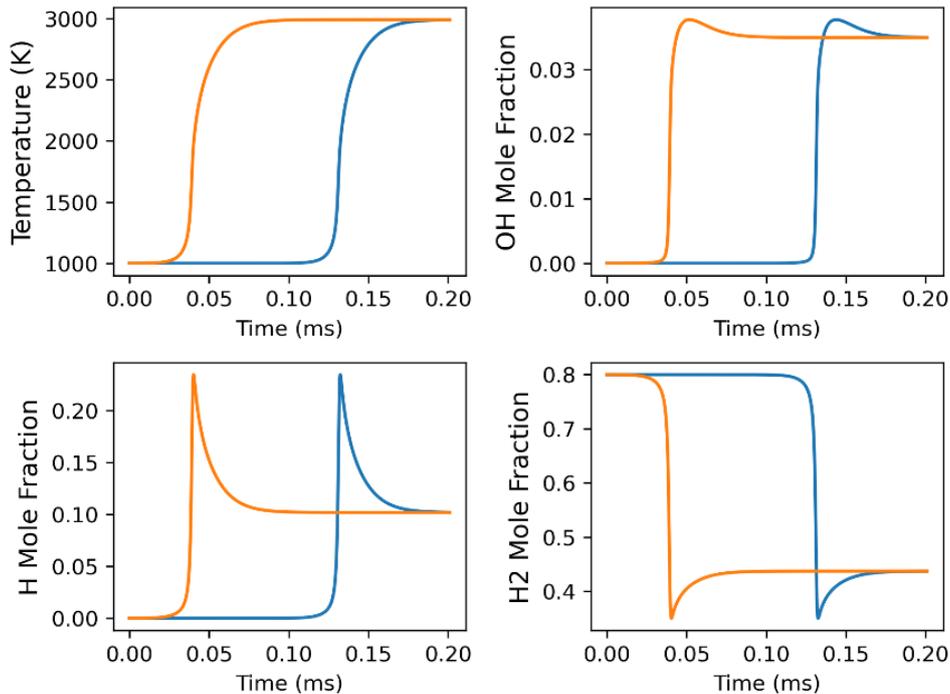


Figure 7. Temperature and species profile of hydrogen/oxygen ignition with and without ozone addition at equivalence ratio $\phi = 2.0$, $P = 1\text{ atm}$ and $T_u = 1000\text{ K}$. (Blue lines: without ozone; orange lines: with ozone)

Next, in order to investigate the effects of equivalence ratio, we examine the situations of lean and rich hydrogen/oxygen mixtures, shown in Figure 6 & Figure 7. For the lean mixture at equivalence ratio of 0.5 shown in Figure 6, there is more oxygen than the fuel could consume; for the rich mixture at equivalence ratio of 2.0 shown in Figure 7, the fuel is abundant, and oxygen can be totally consumed. It is observed that for both situations, the ignition happens faster with ozone additions. For the lean mixture, the ignition delay time changes from 0.075 ms to 0.022 ms. For the rich mixture, the ignition delay time changes from 0.132 ms to 0.040 ms. Both situations show that the addition of ozone could substantially reduce the ignition delay time, which demonstrates that plasma assisted ignition could happen for a wide range of equivalence ratios.

It shall be noted that in addition to hydrogen, methane has also been used as the fuel for rocket propulsion. As such, now we analyze the role of plasma excited species—ozone on the oxidation process of methane.

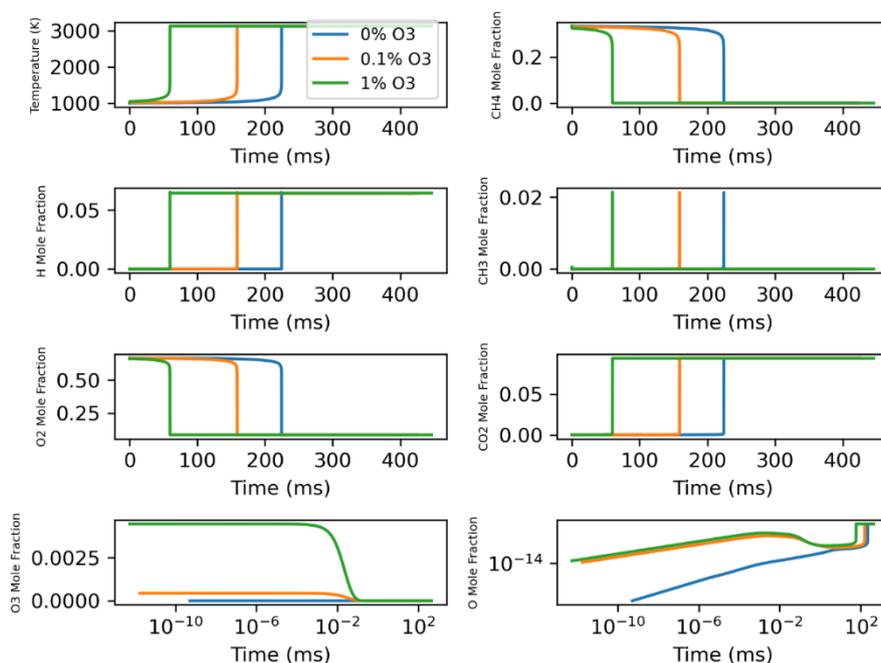


Figure 8. Temperature and species profile of methane/oxygen ignition with different ozone additions at equivalence ratio $\phi = 1.0$, $P = 1 \text{ atm}$ and $T_u = 1000 \text{ K}$.

Figure 8 demonstrates the effect of ozone addition on the ignition process of methane/oxygen mixtures. Similar to the hydrogen/oxygen mixtures, addition of ozone also makes the ignition of methane/oxygen faster. However, with the same percentage of ozone addition (for example, $a = 0.1\%$), the reduction of the ignition delay time for methane/oxygen mixtures is substantially less than that for hydrogen/oxygen mixtures. It is known that the reactivity of methane is lower than that of hydrogen. Therefore, the total ignition delay time for the stoichiometric methane/oxygen mixture is much longer than that for the stoichiometric hydrogen/oxygen mixture. It is thus suggested that plasma assisted ignition is more needed when methane is used as the fuel compared with situations where hydrogen is used as the fuel.

In summary, for the chemical kinetic effects of ozone addition on the ignition processes of both hydrogen/oxygen and methane/oxygen mixtures, the ignition delay times are very sensitive to the addition of ozone. A small amount of ozone addition could substantially reduce the ignition delay time of both hydrogen and methane through the generation of active species such as free radicals, as well as the new reaction pathways activated by the ozone additions. Comparing the two fuels, hydrogen is more sensitive to ozone addition than methane.

4. Plasma Assisted Flame Propagation

In this section, we explore the effect of plasma assistance on the flame propagation process. It shall be noted that the flame propagation is a different phenomenon compared with the ignition process discussed in the previous section. The combustion process starts with the ignition phenomenon, which is the initiation stage of plasma assisted combustion. The ignition process can happen through both the auto-ignition process or the forced ignition by external heating. At the ignition stage, normally the radical concentrations are relatively low, as such the addition of ozone could substantially facilitate the radical generations and make the ignition process faster. Then, the flame propagation is the subsequent steady burning stage of the plasma assisted combustion. In the premixed mode, the flame can propagate in the mixture with certain velocities. Such velocity is considered as an important parameter for quantifying the power generation by burning, and it is also an essential parameter for designing combustion devices in practice. Thus, the current section specifically investigates the plasma assisted flame propagation process.

As a carbon-free fuel, hydrogen has been selected as the fuel for various propulsion systems. The current study first investigates the propagation of plasma assisted hydrogen/oxygen flames. Here, for the propagation of combustion waves, the parameter concerned is the laminar flame speed, which is defined as the propagation speed of the premixed flame in the infinitely large, planar and adiabatic conditions.

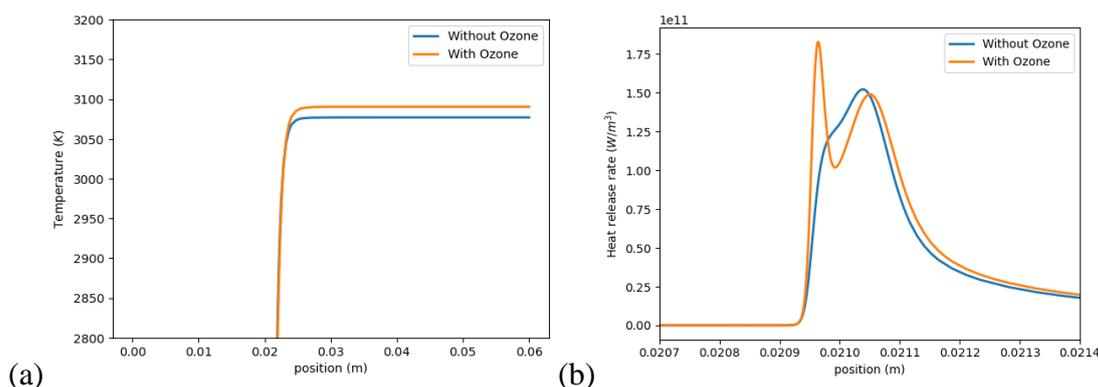


Figure 9. (a) Temperature and (b) heat release rate of stoichiometric hydrogen/oxygen mixtures with and without ozone addition at $P = 1 \text{ atm}$ and $T_u = 300 \text{ K}$.

In Figure 9, the temperature and heat release rate profiles of hydrogen/oxygen mixtures with and without ozone have been demonstrated. For the situation with ozone addition, the amount of ozone added into the mixture is $a = \frac{Y_{O_3}}{Y_{O_2} + Y_{O_3}} = 10\%$. Such ozone additive is substantially more than the situation of ignition. However, in terms of the flame speeds, the case without ozone gives the laminar flame speed of 9.79 m/s and the case with ozone gives the laminar flame speed of 10.83 m/s . As such, the addition of ozone leads to the faster consumption of the fuel in the mixtures. Compared with the ignition process, the flame propagation is less affected by the ozone addition. Therefore, more ozone addition is needed for the situation of enhancing flame propagation compared with the ignition. Additionally, the temperature change due to the addition of ozone is also rather small, with the temperature changed from 3077 K to 3090 K as demonstrated in Figure 9(a). For the heat release rate, one interesting finding is that the case with ozone addition shows two distinct heat release rate peaks; while for the situation without ozone, only one heat release rate peak can be observed (see Figure 9(b)). For the situation of two heat release rate peaks with ozone additions, the peak on the left is due to the ozone decomposition and ozone reactions with other species, and the peak on the right is due to the oxidation of hydrogen. The ozone related chemistry happens at relatively lower temperatures, but the hydrogen oxidation chemistry occurs at relatively higher temperatures. As such the two peaks are separated in space.

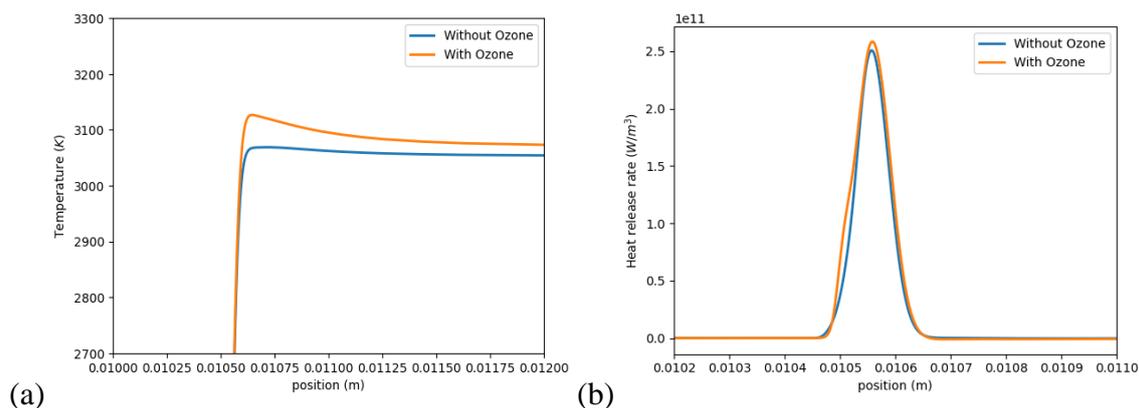


Figure 10. (a) Temperature and (b) heat release rate of stoichiometric methane/oxygen mixtures with and without ozone addition at $P = 1 \text{ atm}$ and $T_u = 300 \text{ K}$.

Next, the effect of ozone addition on the methane/oxygen flame propagation has been demonstrated in Figure 10 for both the temperature and heat release rate distributions. In terms of the enhancement of the laminar flame speed, the speed has been increased from 3.16 m/s to 3.40 m/s , for which the amount of flame speed enhancement is not significant. This finding is similar to the previous results on hydrogen/oxygen flames, which indicates that compared with ignition enhancements, the flame propagation process requires much more ozone additions to increase its propagation speed by plasma excited oxygen. As shown in Figure 10(a), the temperature profile with ozone addition has demonstrated a peak value higher than the equilibrium value at the flame front, and then the temperature decreases to the equilibrium temperature. However, for the situation without ozone addition, no such phenomenon is observed. This phenomenon is also related to the ozone reactions happening near the flame front. As for the heat release rate profiles (see Figure 10(b)), the two situations with and without ozone addition show rather similar results. Different from the situation of hydrogen/oxygen flames, the addition of ozone does not lead to the two-peak phenomenon in the heat release rate profile of methane/oxygen flames.

In summary, the flame propagation phenomenon is less sensitive to the amount of ozone addition into the system compared with the ignition process. The reason behind such observation is partly due to the fact that the initial temperature of the flame is rather low (300 K), and therefore the ozone related reactions cannot be fully activated. Furthermore, for the ignition process, there is no initial radical pool, as such the addition of ozone could significantly facilitate the radical accumulation. However, for the flame propagation, the radical pool is already established. Therefore, much more ozone addition is needed in order to further enhance the radical generation in the flame propagation process.

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