Ignition time improvement in pre-mixture gas of methane, oxygen and argon using plasma

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Effect of plasma on ignition time in pre-mixture methane, oxygen and argon gases was examined using CHEMKIN. Unburned gases has been dissociated by the non-equilibrium plasma, and the ignition time had been calculated for dissociation degree of gases and initial gas temperature. And it is found that the dissociation of oxygen molecules were significantly effective for decomposition of methane. As a result, the first step of combustion process was reacted in advance and the ignition time improved.

1. Introduction

Recently, research on plasma assisted combustion has been done for investigating how to achieve more efficient combustion, more efficient use of fossil fuel and so on. The research is focused on improvements of ignition conditions and combustion conditions and so on by applying electric field or non-equilibrium plasma directly.

Sasaki et al. investigated methane, air premixed gas combustion applying microwave power[1]. While applying the microwave power to the flame, the emission from second positive band system of nitrogen was observed. As a result, acceleration of electrons by microwave electric field has been proved. They also investigated methane, oxygen and argon premixed gas applying dielectric barrier discharge[2]. The emission from excited argon has been observed, and no increase of gas temperature has been measured.

The effect of plasma on the flame has been observed, however, the detail mechanism of applying the discharge or electric field is not cleared, yet. To investigate the effect of non-equilibrium plasma on the flame, the electron transport parameters are calculated[3]. From the calculated electron energy functions, the possibility of existing high energy electrons in the flame, emission of second positive band system of nitrogen molecules and emission from excited argons are confirmed. But the number density of electrons etc. are low enough compared to the densities generated in chemical reactions of the flame.

In this paper, the species which are generated in the plasma could affect the flame or not, are investigated. Especially, the ignition, first stage of combustion is examined.

2. Simulation model and conditions

The simulations have been done using CHEMKIN Pro. The Closed Homogeneous 0D Reactor model is used. The premixed gas ratio is $Ar:CH_4:O_2$ = 0.8:0.05:0.14 [2].

Fig.1 and Fig.2 show the electron collision frequencies in non-equilibrium plasma. In this simulation, to clarify the dissociation effect of oxygen and methane, each dissociated species are examined separately, and the dissociation degrees are estimated using these values.

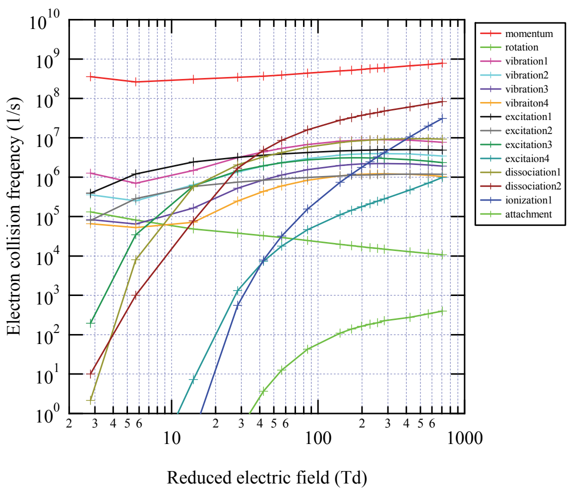


Fig.1 Electron collision frequency of oxygen.

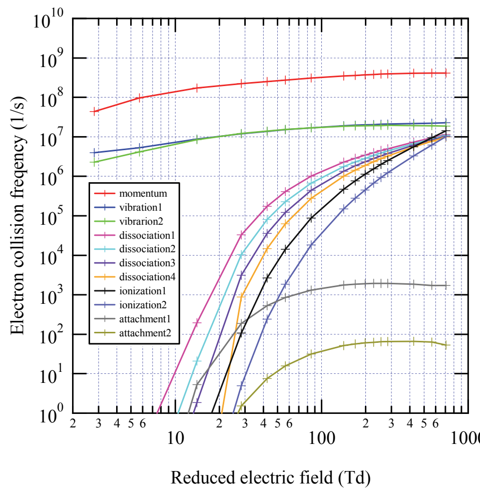


Fig.2 Electron collision frequency of methane.

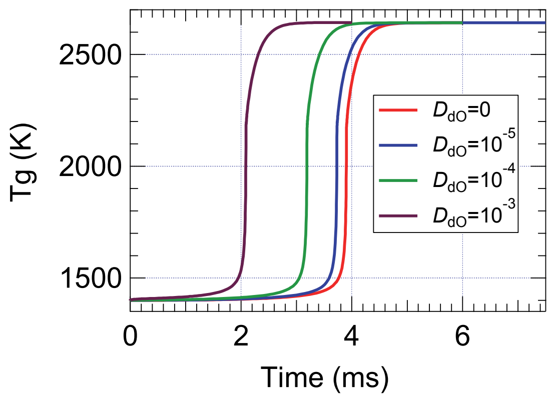


Fig.3 Temporal evolution of gas temperature.

The dissociation degree D_{dO} of oxygen is changed from 0~10^{-3} and the dissociation degree D_{dC} of methane is set to 10^{-5} in the present simulation. And initial gas temperature T_0 is changed from 1400K~2400K.

3. Results and discussions

Fig.3 shows the temporal evolution of gas temperature. As shown in the figure, the gas temperature keeps their values at first, then sudden significant increases happen. At this time, the gases are ignited and start combustion. As the dissociation degree of oxygen increases, the ignition time becomes earlier. In the case of D_{dO}=10^{-3}, the ignition time becomes ~50% compared to that in D_{dO}=0 case. But the gas temperatures after ignition reach to the same temperature in each cases.

In general plasma condition, dissociation rate of oxygen would be 10^{-4}, and methane would be 10^{-5}. At first, to examine the effect of dissociated oxygen atoms, the simulations have been done with oxygen dissociation consideration only. Fig.4 and 5 show temporal evolution of species in mole fraction in D_{dO}=0 case (no dissociation) and D_{dO}=10^{-4} case. Each species fractions are gradually increase and significant change happens at the same time of ignition. In Fig.5, slight differences can be seen at very beginning of the evolutions. These differences would be the effect of dissociated oxygen (~0.8msec). After that, chemical reactions are dominated and the evolutions of species are almost the same in two cases. But the ignition time is shortened in D_{dO}=10^{-4} case.

To investigate early stage of ignition precisely, Fig.4 and Fig.5 are replotted in logarithm for time. Fig.6 and Fig.7 show the temporal evolution of species in fraction mole in D_{dO}=0 case and D_{dO}=10^{-4} case. And early stage of methane combustion pathway is shown in Fig.8. As shown in Fig.6,

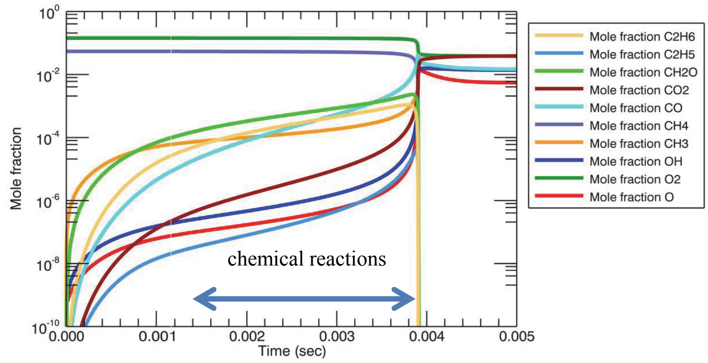


Fig.4 Temporal evolution of species (D_{dO}=0).

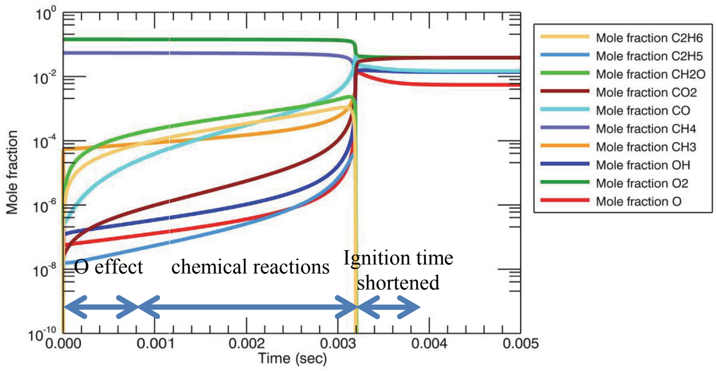


Fig.5 Temporal evolution of species (D_{dO}=10^{-4}).

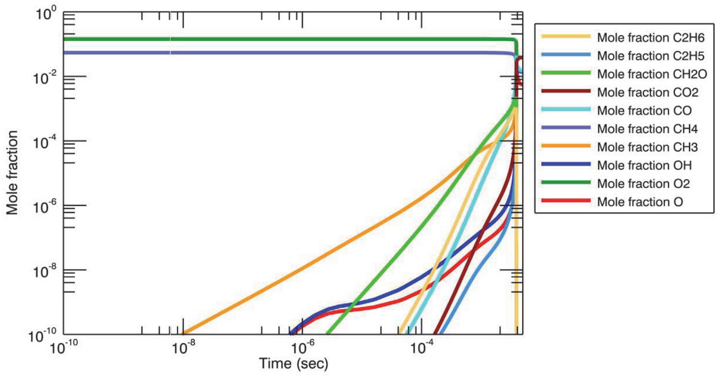


Fig.6 Temporal evolution of species in log scale (D_{dO}=0).

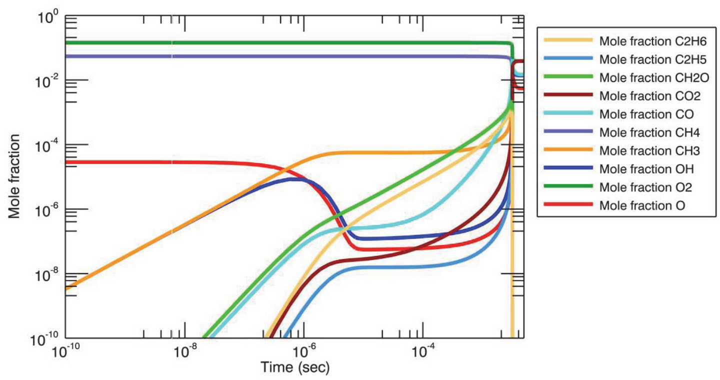


Fig.7 Temporal evolution of species in log scale (D_{dO}=10^{-4}).

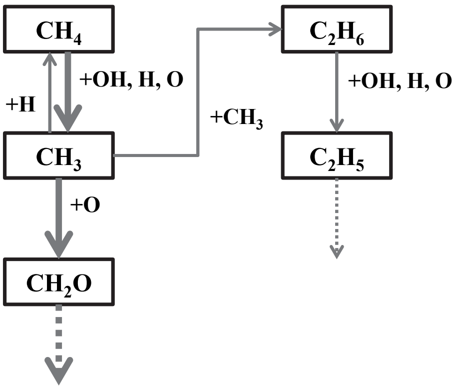
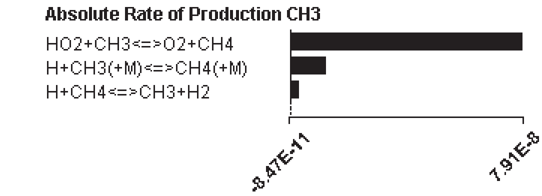
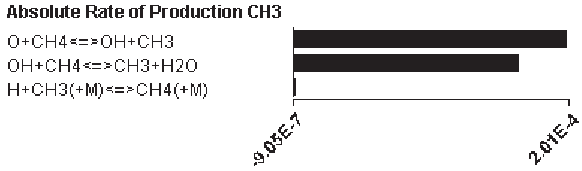


Fig.8 Early stage of methane combustion pathway. (Bold arrows show dominant pathway of combustion)



(a)



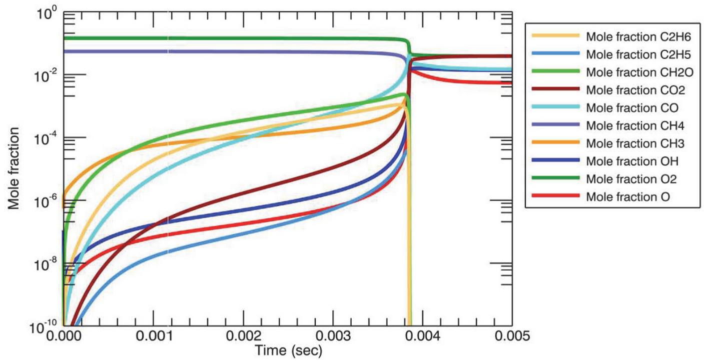
(b)

Fig.9 Production rate of CH_3. (a)D_{dO}=0 case, (b)D_{dO}=10^{-4} case

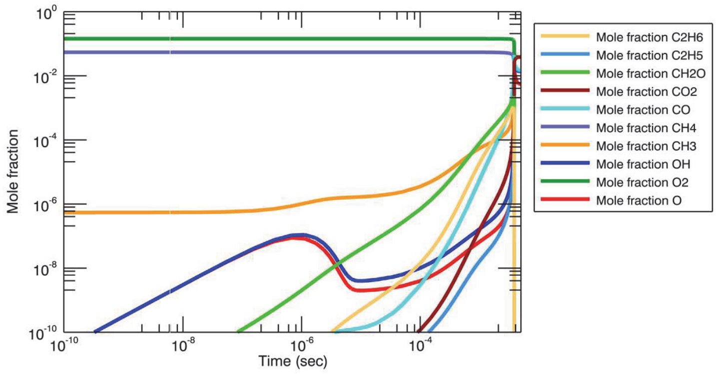
reaction of methane decomposition proceed very slow without plasma. Because the dominant decomposition pathway is reaction with OH, H and O as shown in Fig.8. But in this case methane has to be react with oxygen molecules or other gas species as shown in Fig.9. Every decomposition rate is not large enough to proceed the combustion processes. On the other hand, with plasma of oxygen dissociation rate of 10^{-4}, CH_3 and OH are generated very quickly as shown in Fig.7. One of the major decomposition reaction of methane is reaction with oxygen, and which reaction also generates OH. Another major pathway of decomposition is reaction with OH as shown in Fig.8 and 9. Thus, CH_3 and OH increase in early stage of methane combustion process until O atoms decreases. As a result, double effect of methane decomposition occurs, and other reactions with O and OH proceed.

Fig.10(a) and (b) show temporal evolution of species in mole fraction in D_{dC}=10^{-5} case (methane dissociation case). As shown in Fig.10(a), temporal evolution of species are very similar to those in Fig.4. Because the amount of dissociated methane to CH_3 is very small so the effect of dissociated CH_3 is slight. The ignition time improvement can be seen very slightly. The mole fraction of CH_3 is about 10^{-6} as shown in Fig.10(b). This value is almost 1/100 value obtained in the case of D_{dO}=10^{-4} (oxygen dissociation case) as shown in Fig.7. O atoms and OH molecules are also obtained as the processes proceed, but the amount of mole fraction is small. As a result, the dissociation of methane only is not so effective at this dissociation degree.

Fig.11(a) and (b) show temporal evolution of species in mole fraction in D_{dO}=10^{-4} and D_{dC}=10^{-5} case. As mentioned in above, there is less effect of methane dissociation. Until time is earlier than 1msec, there are slight differences can be seen between



(a)



(b)

Fig.10 Temporal evolution of species (D_{dC}=10^{-5}). (a)time in linear scale (b)time in log scale

(CH_3+O) case and (O only) case. But after that, the evolutions of species are almost the same. And the ignition time improvement can be seen slightly. As a result, there is no synergy effect of CH_3 and O, there is significant O dissociation effect in this conditions. But more wide range parametric study of methane dissociation degree would be needed.

Fig.12 shows the ignition time delay dependence. Red line shows typical ignition delay dependence on initial gas temperature. The ignition time deay linearly decreases with increasing initial gas temperature as shown in the figure. And as dissociation degree of oxygen increases, ignition time delay decreases (becomes earlier) at the same initial gas temperature in lower initial gas temperature region. There is an improvement of ignition time in this region. But in high initial gas temperature region, the effect of oxygen dissociation becomes weaker. On the other hand, no improvement of ignition time is require, the initial gas temperature can be lowered. This result leads to less gas heating or less pressuring in combustion system.

4. Conclusion

The effect of specie dissociated by plasma is examined using CHEMKIN Pro. Most effective species is oxygen atom, but CH_3 molecules are slightly effective in the present condition. There is no synergy effect of O and CH_3 molecules.

Dependence of dissociation effect on initial gas temperature is examined. The effect of dissociation of oxygen is effective in lower initial gas temperature condition. But in high temperature region, the effect becomes vanished.

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References

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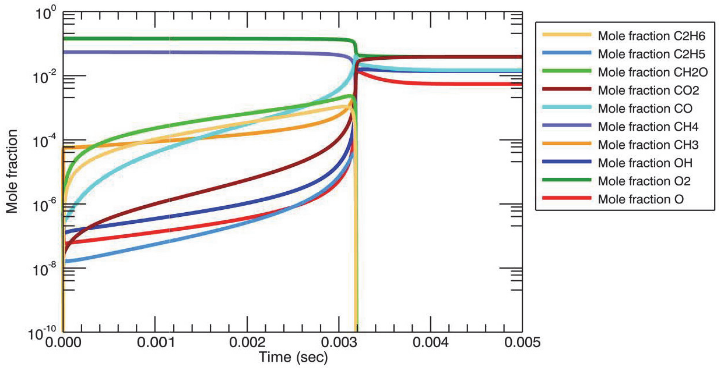
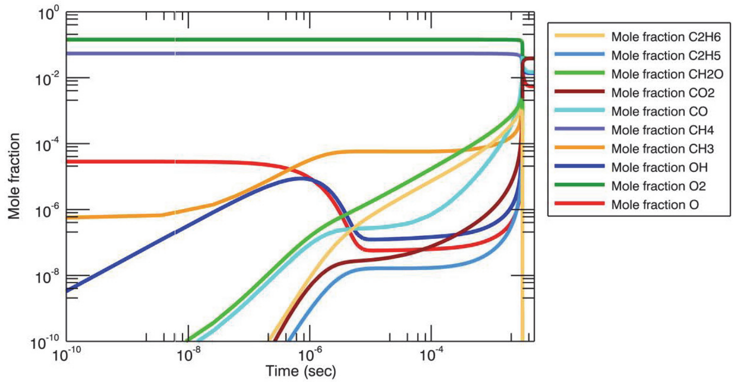


(a)



(b)

Fig.11 Temporal evolution of species (D_{dO}=10^{-4}, D_{dC}=10^{-5}). (a)time in linear scale (b)time in log scale

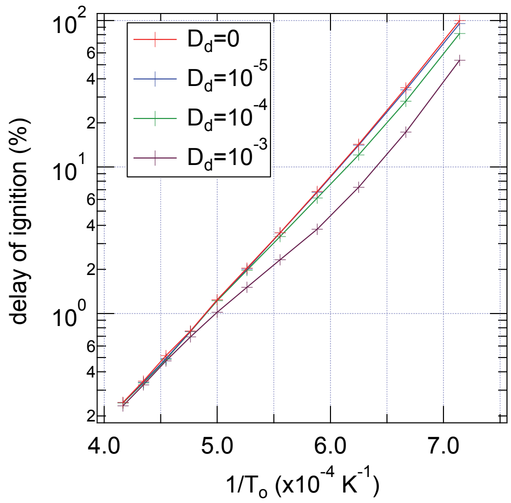


Fig.12 Ignition time delay dependence on initial gas temperature and dissociation degree of oxygen.