EFFECTS OF ETHANOL ADDITION ON SOOT PARTICLES DYNAMIC EVOLUTION IN ETHYLENE/AIR LAMINAR PREMIXED FLAME

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This work focuses on the effects of ethanol addition on soot particle size distributions functions (PSDF) in ethylene/air premixed flame. The mixed ethanol/ethylene chemical reaction mechanism was used to calculate the concentrations of gaseous components in the premixed flame. A Monte-Carlo stochastic method was used to solve the particles dynamical model, which including particle inception, coagulation and surface reaction. PSDF in different heights of the flame were obtained with different amounts of ethanol added into ethylene. Three different equivalence ratios (1.78, 2.01 and 2.34) and four different blend ratios of ethanol/ethylene (0%, 10%, 20%, and 30%) were investigated. The simulated results showed that mole concentrations of some important oxidant species, i.e. OH, H and O, were enhanced by ethanol addition. Thus, the soot and soot precursor's formation, such as C_2H_2, PAHs, etc., were restricted owing to the increasing oxidation reaction with ethanol addition. Meanwhile, large particles were reduced when ethanol blended to the ethylene at the same height of the flame.
Key words*: ethanol addition; soot formation; particle size distribution; premixed flame*

1. Introduction

Oxygenate additives doped into fuels are considered as an effective way to reduce both of the soot and nitrogen oxides (NOx) [1-6]. Methanol (CH_3OH), ethanol (C_2H_5OH) and dimethyl ether (CH_3OCH_3) have been widely used to reduce the engine emissions. Many researchers have also studied the effects of ethanol addition on pollutant emissions theoretically and experimentally, such as NOx, CO_2 and soot [7-8]. Their results showed that ethanol addition can effectively reduce the particulate matters (PM). Lapuerta et al. [7] found that bioethanol reducing soot emissions without increasing other gaseous emissions by the engine test. Chen et al. [8] also researched the effects of ethanol addition on smoke and PM with a diesel engine. They measured the total mass of the particles emitted from the tested engine and found that the addition of ethanol could significantly reduce the PM.

Many researchers also studied the influences of ethanol addition in ethylene (C_2H_4) or benzene flame in order to understand the mechanism of the oxygenated additives suppressing soot formation. In earlier studies, Frenklach and co-workers [9] found that both ethanol and methanol can reduce soot formation, and ethanol is

more effective. However, Alexiou and Williams [10] found that methanol is more effective than ethanol in suppressing soot production. They considered that ethanol produces more acetylene, which is an important species for soot growth. Wu et al. [11] tried to explain the influence of ethanol addition by studying the combustion process in a premixed ethylene flame. They measured the polycyclic aromatic hydrocarbons (PAHs) concentration in the investigated flames and found that ethanol addition can reduce the PAHs (two and three rings) concentration. In the recent experimental study of ethylene premixed flames, the effects of ethanol addition on soot particle size distributions functions (PSDF) were obtained by Salamanca et al [12]. They found that the addition of ethanol mostly affects the gas phase chemistry and slowdown the formation of soot precursor and soot. But due to the limitation of experimental resources, they did not give a more detailed explanation.

However, most of the previous studies only researched the effects of ethanol addition on the total amounts of soot particles. Moreover, most of them were based on experiments and gave little attention on the PSDF. Recent studies [13-14] shown that PSDF is important because particle radiative property, optical property and toxicity are closely related with particle size and number density. Meanwhile, new traffic laws in many countries not only limited the soot mass but also the particle quantity. In real combustion process, variation of some flame environment, such as temperature and equivalence ratio, can change PSDF because the surface growth and oxidation processes can be promoted or inhibited [15-16]. Though the influence of equivalence ratio on PSDF of ethanol/ethylene/air flame has experimentally investigated by several researchers [12], there is still little knowledge about the variations of PSDF in complicated combustion process of blended fuels.

This work researched the influences of ethanol addition on the particle size distribution and total soot mass in the ethylene/air flame. The pure ethylene set as the baseline flame, three different amounts of ethanol addition were investigated, 10%, 20% and 30%, respectively. Three fuel-air equivalence ratios were proposed, 1.78, 2.01 and 2.34, respectively. Particle size distributions in different HAB (height above the burner) were calculated by using the detailed soot model, including the detailed chemical kinetic reaction and particle dynamical process. Monte Carlo stochastic method was used to solve particle dynamical model, which contained four different sub-processes: particle nucleation, particle coagulation, gaseous species growth and particle oxidation.

2. Computational details

The simulation of PSDF in the present work was divided into two steps. Firstly, the flame structure, gaseous specie concentrations and soot moments [17-19] in the premixed flame were calculated using a PREMIX computer code [20] and a detailed chemical mechanism. The PREMIX computer code had been coupled with a soot model [21, 22], and the detailed chemical mechanism described the oxidation of the ethylene/ethanol fuel and the formation of PAHs. Secondly, the PSDF was calculated with a Monte Carlo stochastic approach using results from the first step as an input.

2.1 Chemical reaction kinetics model

The chemical mechanism of mixed ethylene/ethanol fuels was built by Ilya E. Gerasimov and co-workers [23]. The mechanism included 121 gaseous species and 708 elementary reactions, which contained two major parts: the ethylene oxidation mechanism from the Ref. [18,19], and the oxidation mechanism of ethanol was developed by Marinov [24]. The mechanism includes the oxidation of ethylene/ethanol fuels, formation and

oxidation of C1-C6 species, and PAHs formation and oxidation. The maximum ring of PAHs is up to four in this mechanism.

The PAHs growth (formation of naphthalene (A_2) to pyrene (A_4) in this study) was described by the hydrogen abstraction acetylene addition (HACA) mechanism after the first ring formed [18], as shown in (R1) and (R2):

$$PAH_i + H \rightarrow PAH_i\cdot + H_2, \tag{R1}$$

$$PAH_i\cdot + C_2H_2 \rightarrow PAH_{i+1} + H\ . \tag{R2}$$

PAHs oxidation was described by the following reactions,

$$PAH_i\cdot + O_2 \rightarrow PAH_{i-1} + products, \tag{R3}$$

$$PAH_i + OH \rightarrow PAH_{i-1} + products. \tag{R4}$$

2.2 Particle dynamical model

Particles dynamical model includes nucleation, particle coagulation, and surface growth. Surface growth includes the PAHs condensation and acetylene growth on the surface by HACA mechanism. Meanwhile, the oxidation of particles by OH and O_2 was also considered [25]. The PSDF can be described by the following equation:

$$\frac{\partial N_i}{\partial t} = R(t)\delta_{in} + G(t,\ i) + W(t,\ i), \tag{1}$$

where N_i is the number density of particle i, $R(t)$ is the nucleation rate of primary particle, δ_{in} is the smallest particle in the present study, $G(t,i)$ is the coagulation rate, and $W(t,i)$ is the surface reaction rate of particles.

In the present study, soot inception was described by the dimerization process of two A_4 molecules:

$$A_4 + A_4 \rightarrow Soot, \tag{R5}$$

$$R(t) = \frac{1}{2}\beta N_{A4} N_{A4}, \tag{R6}$$

where, $R(t)$ is nucleation rate, β is the collision kernel, and N_{A4} is the mole concentration of A_4.

Soot particles coagulation process can be described by Smoluchowski's coagulation equation [17] as follows:

$$G(t,i) = \frac{1}{2}\sum_{j=1}^{k-1}\beta(t,i-j,j)N(t,i-j)N(t,j) - \sum_{j=1}^{n_{\max}}\beta(t,i,j)N(t,i)N(t,j), \tag{2}$$

where $\beta_{i,j}$ is the coagulation frequency.

Soot surface growth reaction includes A_4 condensation on the soot surface and acetylene (C_2H_2) addition through the HACA mechanism. In this work, soot oxidation was considered as reactions with two gaseous species, O_2 and OH. The soot growth through the surface reactions mechanism was described using R7-R10:

$$Soot_i + A_4 \rightarrow Soot_{i+16}, \tag{R7}$$

$$Soot_i + C_2H_2 \rightarrow Soot_{i+2}, \tag{R8}$$

$$Soot_i + OH \rightarrow Soot_{i-1}, \tag{R9}$$

$$Soot_i + O_2 \rightarrow Soot_{i-2} \cdot \quad \text{(R10)}$$

The above reactions (R7-R10) can be described by the following equation:

$$W(i) = \sum_{l=1}^{4} [w^l(i-\delta^l{}_i)N(i-\delta^l{}_i) - w^l(i)N(i)], \quad (3)$$

where $w^l(i)$ represents the rate of surface process l, and $\delta^l{}_i$ is the addition or reduction of carbon number caused by the process l. The definition of the process l are as follows: A_4 condensation, $\delta^l{}_i$ =16; C_2H_2 growth, $\delta^l{}_i$ =2; oxidation reaction by O_2, $\delta^l{}_i$ =-2; oxidation reaction by OH, $\delta^l{}_i$ =-1. The calculations of $w^l(i)$ are developed by Frenklach et al. [18,19].

In solving Eq. (1), the evolution of particles dynamic with time can be represented by a stochastic particle system. Tracking every soot particle is impossible in currently computational conditions because the particle number is too large. In this study, one randomly generated particle represented a specified number of particles, which could be different for each calculation. Temperature profiles and species concentrations were obtained from the PREMIX flame simulation. The profiles were listed as function of residence time. By using these rates of the particle dynamic events mentioned above, the time step was calculated according to the exponentially distributed random variable, which were explained carefully in Ref. [26]. Subsequently, one event was chosen probabilistically to happen according to the rates of the events. After completing a time step, the whole particles in the system were updated. Meanwhile, a new time step was determined. The Monte-Carlo stochastic method was derived from a procedure called sweep [27] and the accuracy and numerical efficiency had been discussed in Ref. [26]. The whole size distribution of soot particles was resolved by this stochastic approach and efficient computation.

3. Model validation

In order to validate the model built in present work, calculations were performed and compared to experimental results previously obtained in the literature [23]. The calculation conditions were in corporate with the experimental conditions. The temperature profiles used were obtained from measured results.

Fig. 1 presents comparisons of results between the model and experiment for some gaseous species, such as CH_4, C_2H_2, H_2O and CO_2 et al. As seen from Fig. 1, the experimental results and simulation results both indicate that 50% ethanol addition to the ethylene/air mixture cause a change of the composition in the intermediate and final products, i.e., an obviously increase in the mole fractions of CO_2, H_2O, H_2 and a decrease in the CO and C_2H_2 mole fraction. Some of these species (H_2, O_2, CO_2) are matched well using the present model. The mole fraction of C_3H_3 is not matched well and the calculated result is a little higher than the experimental data, but the deviation is still within the acceptable range. Meanwhile, the mole fractions of CO are not matched well too. The maximum deviation between the calculated data and experimental data almost reached to 14%. This can be due to the products which containing carbon other than CO and CO_2 were not taken into account at the higher HABs. The mole fractions of C_2H_2 are also not matched well at higher HAB, this may be explained by the absence of the consumption of C_2H_2 at the higher HAB regions.

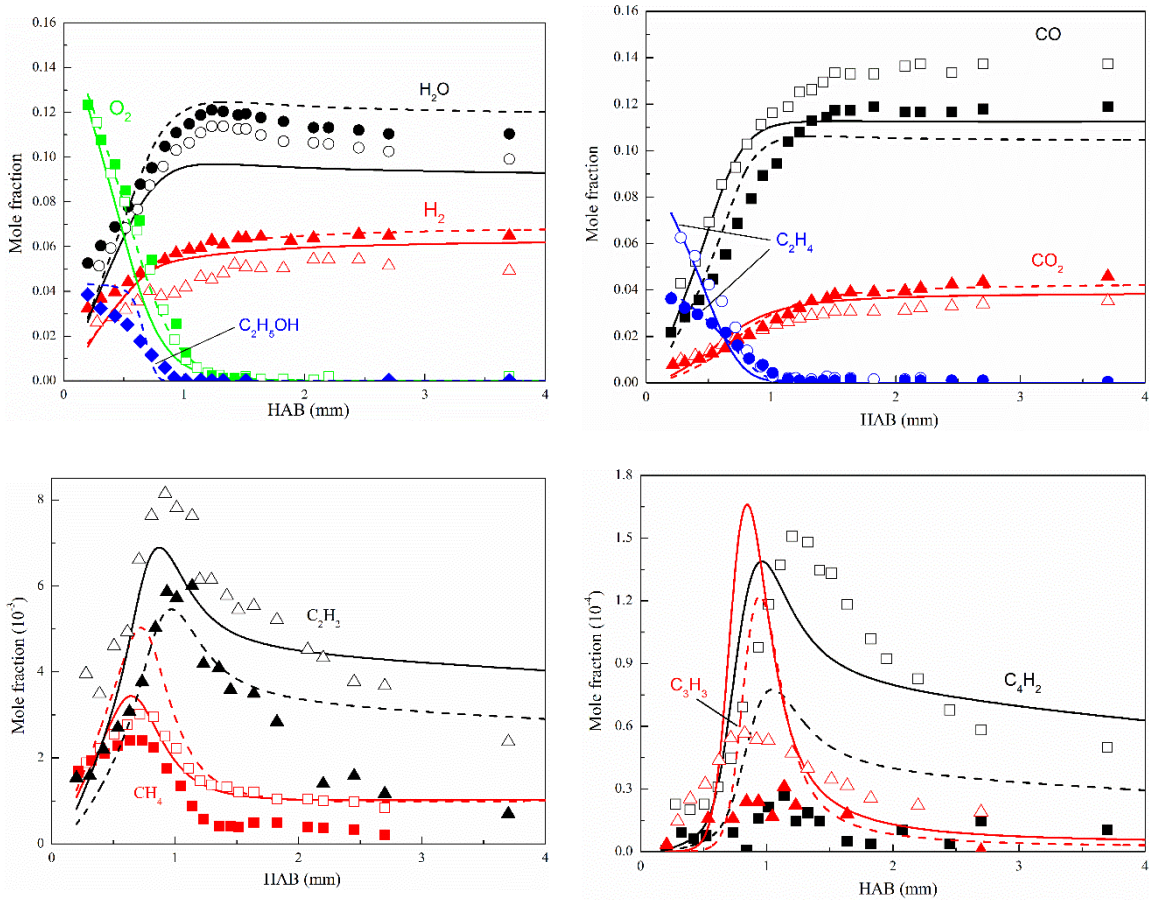

Fig. 1 Comparison of measured [23] (symbols) and calculated (lines) some species concentrations. Open symbols and solid lines represent the species concentrations in C_2H_4 flame, filled symbols and dashed lines represent the species concentrations in C_2H_4/ethanol flame

In order to validate the soot model built in present work further, the soot volume fraction in ethylene premixed flame was also simulated to compare with experimental measured results [28], as shown in Fig.2. The initial conditions were consistent with the experimental conditions in Ref. [28]. EQMOM (Extended Quadrature Method of Moments) model results [29] were also described in Fig.2 to compare with our work's results. It can be observed that the present soot model is able to reproduce the soot volume fraction fv at lower HABs of C2H4 premixed flame, but under-predict the fv at higher HABs compared to EQMOM model. It should be noticed that the models used in this work did not take into account soot aggregation but the EQMOM simulations did. In addition, it is well known that the ethylene oxidation mechanism [18-19] under-predict the PAH concentrations, which are important precursors for soot formation, by order of magnitudes [30-31]. This can also be the root cause of the discrepancy between calculated and experimental values. The difference between the experimental and simulation results is considered to be acceptable. In this work, the simulation results of Fig.1-2 showed that the model built in this work can capture the major characteristics of the flame structure and soot formation, which can be used for the prediction of soot formation.

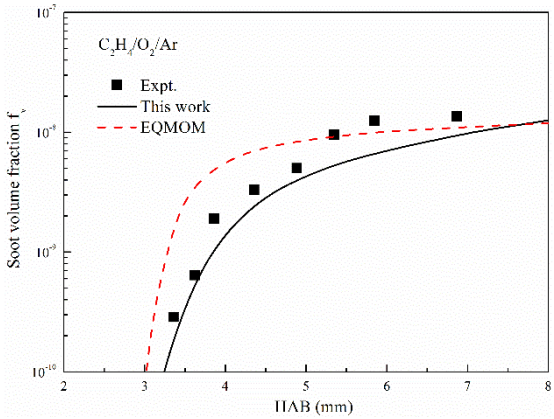

Fig. 2 Comparison of soot volume fraction for a premixed C_2H_4/O_2/Ar flame from this work's results (solid line), EQMOM model [28] (dashed line) and experimental results [29] (symbols)

4. Results and discussion

The concentrations of H, H_2, C_2H_2, O_2, OH, H_2O, and A_4 were used as the input to the soot model that described particle dynamical processes. This soot model includes nucleation, particles coagulation, surface growth and oxidation, which constructed by Frenklach and Wang [19]. Monte Carlo method is used to solve this soot model and calculate the PSDFs in four different flames (with 0%, 10%, 20% and 30% ethanol addition in C_2H_4 fuel). These calculations were performed at the atmospheric pressure, the temperature of mixture gas was set at 423K, and the equivalence ratios were set 1.78, 2.01 and 2.34, respectively.

The mole fraction of major gas species are plotted in Fig.1. There are two important pathways of ethanol dissociation:

$$CH_3CH_2OH = C_2H_4 + H_2O, \quad \text{(R11)}$$

$$CH_3CH_2OH = CH_3 + CH_2OH. \quad \text{(R12)}$$

The mole concentrations of H, O, OH and H_2O are increased by the addition of ethanol (see Fig.3). The calculated peak mole fractions of H, OH and H_2O increased by 9.4%, 8.8%, and 19.2%, respectively, compared with those in the pure ethylene flame. This can be explained by reactions R11 and R13, i.e. ethanol addition caused more H_2O and OH.

C_2H_2 is known as important PAH precursor and key species for soot formation. By analyzing the final concentration of acetylene, it is noticed that the concentration of C_2H_2 is decreased when ethanol doped in the flame (see Fig.1 and Fig.3). This is due to the presence of the ethanol which enhances the formation of OH radical, and accelerates the consumption of acetylene (R14).

$$H_2O + H = OH + H_2, \quad \text{(R13)}$$

$$C_2H_2 + OH = CH_2CO + H. \quad \text{(R14)}$$

The R15 is the recombination of two C3H3, which is the most important pathway to form benzene (A_1). According to the research of Ref. [32], this pathway represents 70% of A_1 formation. The second important pathway is the H addition reaction from phenyl (A_1-) to forms A_1, which accounts for 27% of A_1 formation [32]. The C_3H_3 concentration decreased with the reduction of C_2H_2 concentration and the rise of OH concentration. According to R16 and R17, the formation of C_3H_3 is largely depended on the mole fraction of C1 and C2 species, such as C_2H_2 CH_2 and CH_2*. With the increase of OH and decrease of C_2H_2 and C_3H_3, the A_1 is decreased.

$$C_3H_3 + C_3H_3 = A_{1,} \quad \text{(R15)}$$

$$C_2H_2 + CH_2 = C_3H_3 + H, \quad \text{(R16)}$$

$$C_2H_2 + CH_2* = C_3H_3, \quad \text{(R17)}$$

$$C_3H_3 + OH = CH_2CHCHO, \quad \text{(R18)}$$

$$C_3H_3 + OH = C_2H_3 + HCO. \quad \text{(R19)}$$

H atom plays an important role in PAHs growth, which can be described by reactions R20-R23. Where A_i represents PAHs including the number of aromatic rings i, A_i- is the corresponding aromatic radical, and $A_iC_2H_2$- is formed by the addition of C_2H_2 to A_i-. The aromatic molecules become to radicals by the H atoms and the PAHs growth process is promoted. However, it can also be observed that the reaction of O_2 and H produced more OH and O (R20), thus oxidation reaction of PAHs enhanced. The lower C_2H_2 concentration results in a lower PAHs production rate. Meanwhile, the aromatic radicals are restricted by the abundance of H resulting in a lower production rate of larger PAHs.

$$O_2+H=O+OH, \quad \text{(R20)}$$

$$A_i + H = A_i- + H_{2,} \quad \text{(R21)}$$

$$A_i- + C_2H_2 = AiC_2H_2-, \quad \text{(R22)}$$

$$A_iC_2H_2- + C_2H_2 = A_{i+1}+H. \quad \text{(R23)}$$

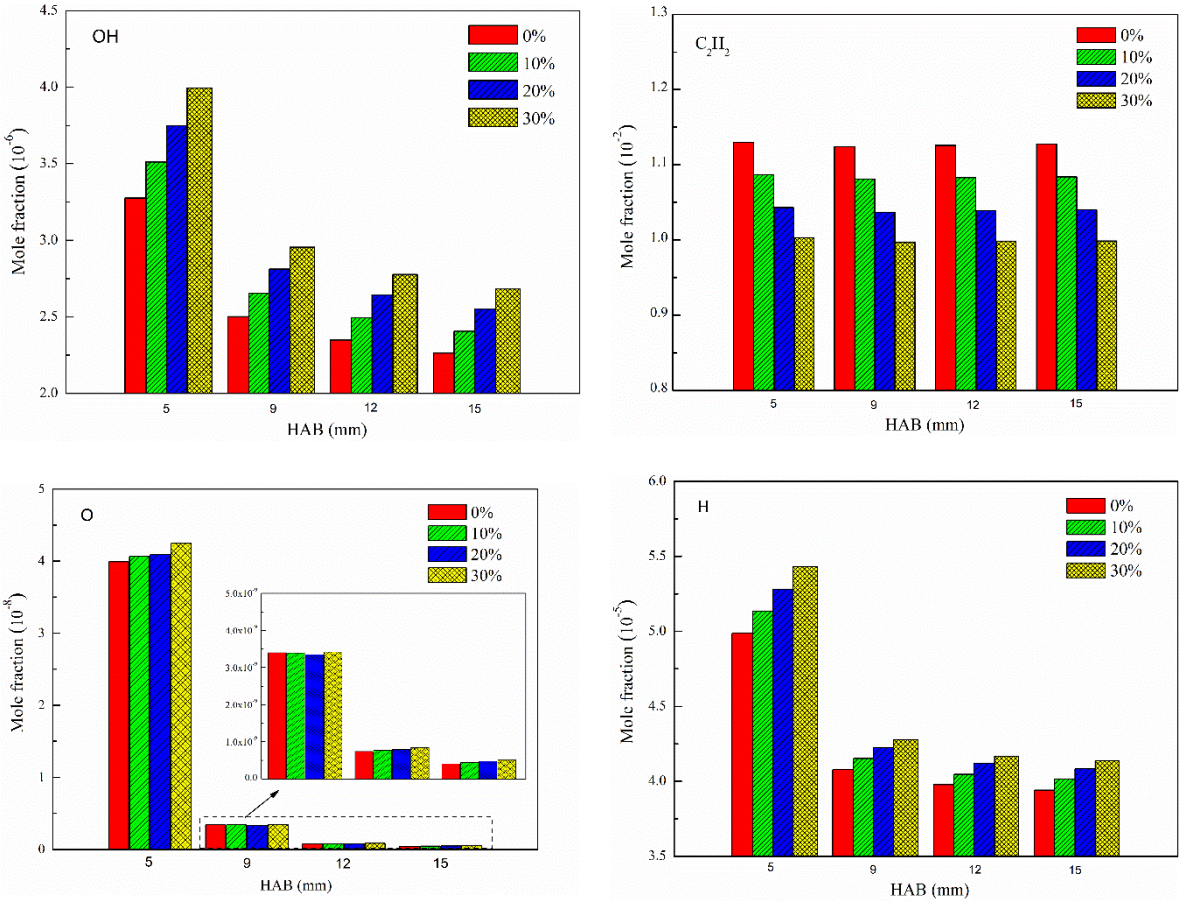

Fig. 3 Mole fraction profiles of OH, C_2H_2, O and H with different amounts of ethanol

The mole fraction profiles of A_1 and A_4 are presented in Fig. 4, in which the latter are believed to be the soot inception species. We can clearly see reduction of the A_1 and A_4 formed in flames when ethanol is added. The peak of A1 and A4 mole fraction decreased 25.3% and 28.9%, respectively, when 30% ethanol added.

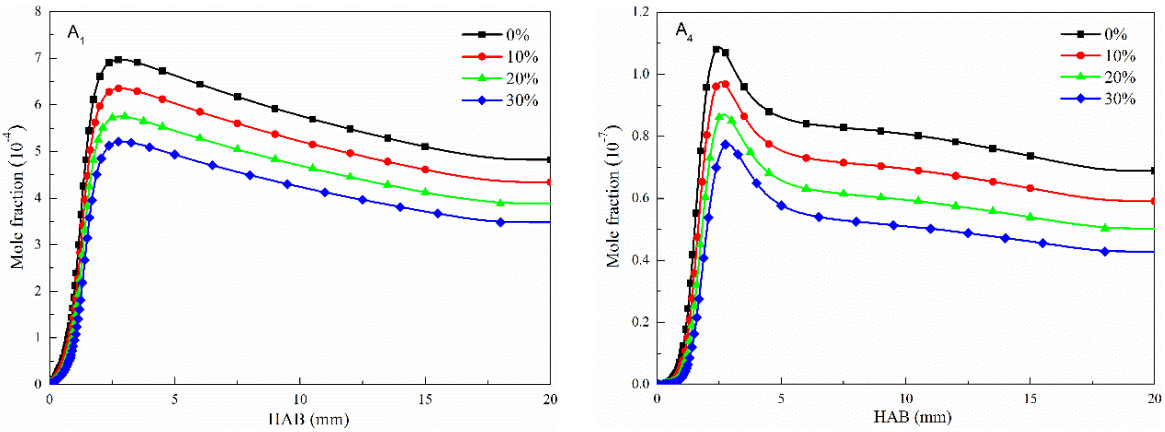

Fig. 4 Mole fraction profiles of A_1 and A_4 with different amounts of ethanol at an equivalence ratio of 2.01

The effect of ethanol addition on larger aromatics should be similar to those on A_1, and the results consistent with Wu's measurements [11]. The lower A_1 concentration combined with lower C_2H_2 levels slowed down the PAHs growth corresponding to the HACA mechanism. PAHs and C_2H_2 can directly affect the soot inception and surface growth reaction, thus soot formation decreased.

The soot mass concentrations at HAB=12mm with ethanol addition at different equivalence ratios, which are calculated by the method of moment, are described in Fig. 5. Soot mass concentrations are reduced with ethanol addition in the presented three equivalence ratios. The same results can also been found in Wu and co-workers' research [11]. Another important finding is that the reduction percentage of soot mass at the equivalence ratio of 1.78 is highest among three equivalence ratios with the same amount of ethanol addition. The reduction percentages of soot mass with 10%, 20% and 30% ethanol addition are 21.34%, 39.95% and 54.62%, respectively, at the equivalence ratio of 1.78. This is because the soot precursors is reduced more and soot particles oxidation is more severely with ethanol addition in lower equivalence ratio.

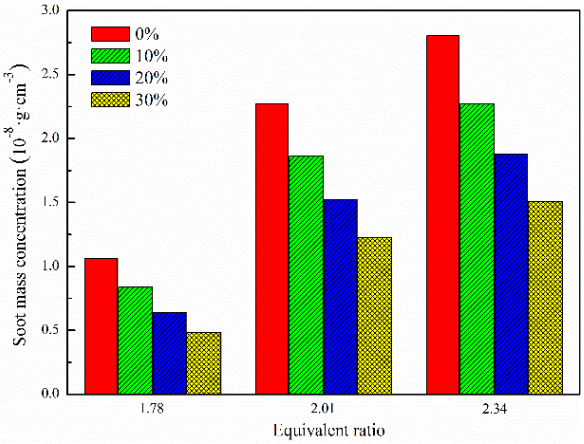

Fig. 5 Effects on soot mass concentration with different amounts of ethanol

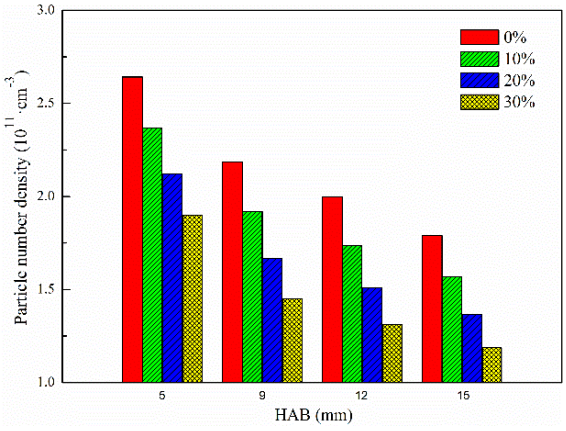

Fig. 6 Effects on particle number density with different amounts of ethanol

Fig. 6 presents the soot particles number density obtained for all flames of the equivalence ratio of 2.01 at HAB = 5, 9, 12 and 15 mm, respectively. Soot number density mainly affected by the nucleation species (i.e. A_4) concentration, so the variation of soot number density is accordance with the variation of A_4 concentration: soot number density decreased with increase of the ethanol mole fraction in ethylene flame.

Fig. 7 presents the PSDF obtained for all flames of the equivalence ratio 2.01 at HAB = 5, 9, 12 and 15 mm, respectively. For all flames, the average diameter of soot particles increases with the HAB rises. Results of these flames represent a typical characteristic of PSDF in premixed flame: at the lower HAB, a unimodal characteristic can be seen, with the HAB increase, a bimodal behavior start to appear. The first mode is characterized by a high number concentration of small size particles, which slightly decreases when HAB increases. The small particles turned into large particles by two important pathways: coagulation and surface growth.

When ethanol added into the ethylene flame, the unimodal distribution also can be seen in the lower HAB. Meanwhile, other two important finds are observed with the ethanol content increased: firstly, the particle number density in the first modal slightly decreased and the second modal is hardly to formation at lower HABs. Secondly, the number of large size particles is reduced with ethanol and the peak of larger particles is putting forward. In the pure ethylene flame, the bimodal distribution is already obvious at 9 mm above the burner. When the content of ethanol addition goes to 30% in the flame, PSDF made the following changes: the position of bimodality appeared is putting forward, at about 12 mm above the burner, and particles are becoming smaller, which are not larger than 15 nm. Owing to the reduction of soot precursors species, such as C_2H_2 and A_4, the particle nucleation and surface growth processes are suppressed in ethanol/ethylene/air flame. Meanwhile, the soot oxidation reaction is enhanced because of the oxidative radicals (O, OH, et al) increased. As a result, the final soot size is restricted and the second modal position of PSDF is putting forward.

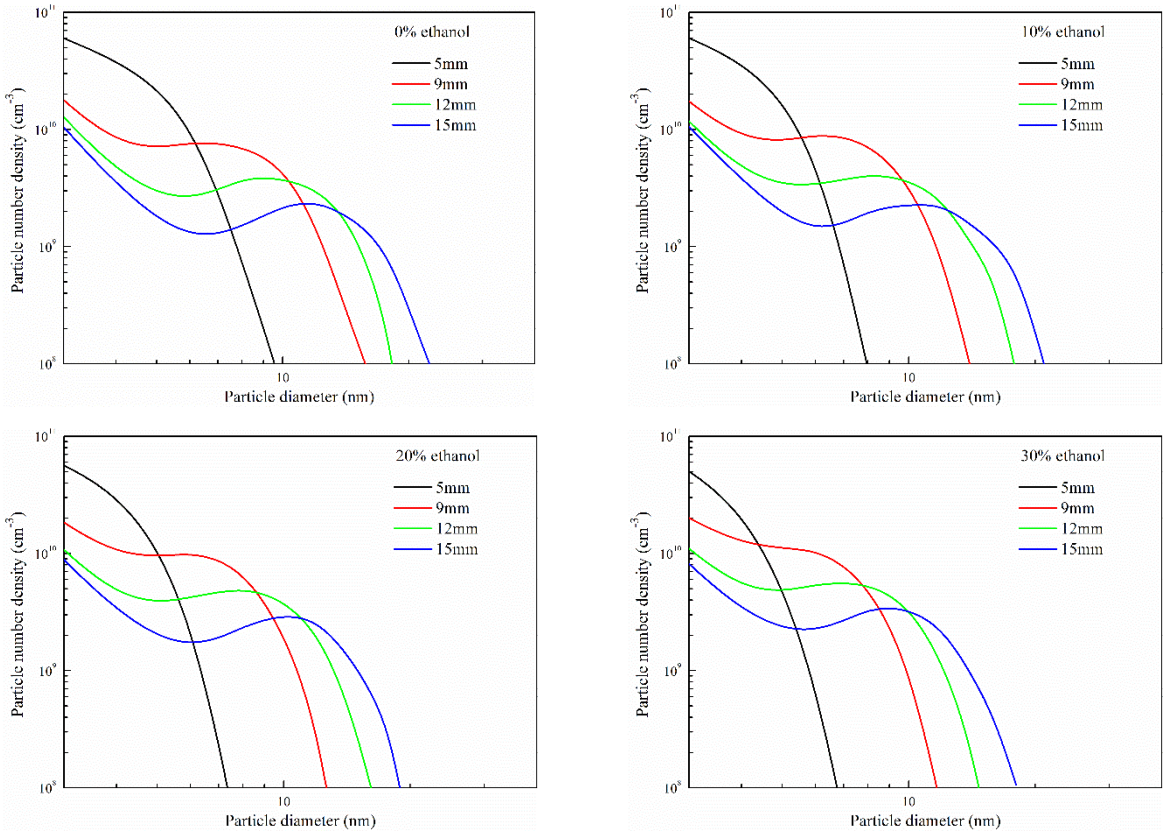

Fig. 7 Particle size distribution obtained for all flames at an equivalence ratio of 2.01

Fig. 8 reports the PSDF obtained for the different flames at three equivalence ratios, 1.78, 2.01 and 2.34, respectively. We can clearly see that the number of small size particles and the maximum particle size are reduced significantly at lower equivalence ratio. It can also be observed that the bimodal characteristic becomes insignificant at lower equivalence ratio under the same HAB. There is almost no bimodal characteristic at the upper regions (HAB=9 and 12 mm) at lower equivalence ratio. As stated before, the small particles turned into large particles by particle coagulation and surface growth. At a constant equivalence ratio, the small particles number is reduced and larger particles are produced under higher HAB, which can largely due to the coagulation process. Ethanol addition restricts the particle nucleation and surface growth, enhances the particles oxidation, thus the final particle size is small (Fig. 9). Particle size distribution evolution is the combined effects of nucleation, particle coagulation, surface growth and oxidation.

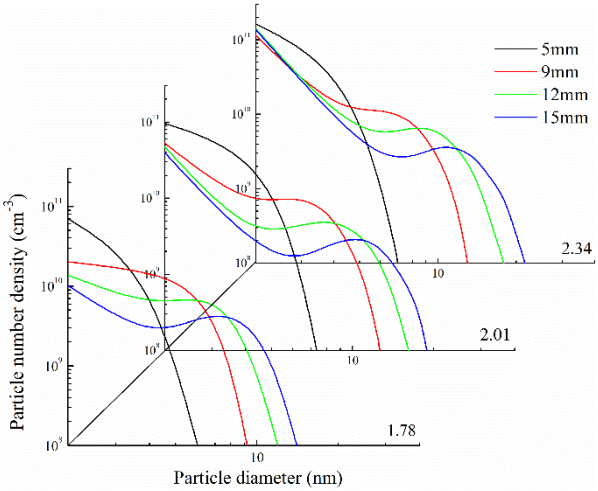

Fig. 8 Particle size distribution for ethylene/air flames at different equivalence ratios

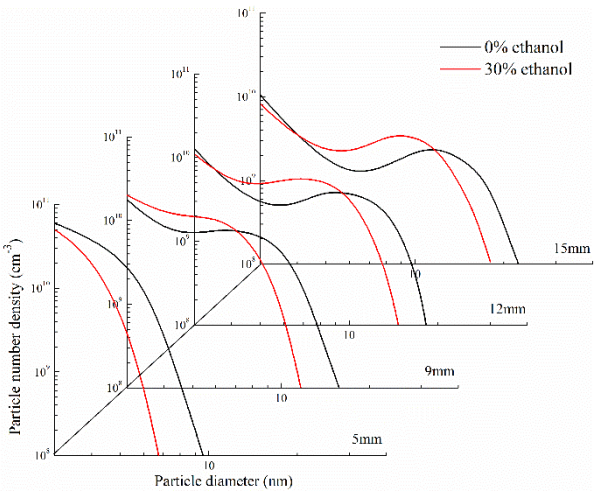

Fig. 9 Particle size distribution obtained at an equivalence ratio of 2.01

5. Conclusion

In this paper, the influence of ethanol addition on soot and gas products in premixed ethylene/air flame had been analyzed. A detailed soot model had been built, which includes detailed chemical mechanism and particle dynamical model. Monte Carlo method was used to solve particle dynamical model. Flame conditions with four different ethanol added contents and three different equivalence ratios were investigated, particle size distributions were obtained. The results showed that the addition of 10%–30% of ethanol in ethylene effectively reduced the concentrations of soot precursors, soot number density and the total mass of soot. The concentrations of H, O and OH were increased, and the soot precursors and soot oxidation rates were also increased. By comparing the soot reduction percentages in three different equivalence ratios, we found that the suppression of soot formation could be more effectively at lower equivalence ratio by ethanol addition. These results are in good agreement with the main trends observed in experimental tests [32]. The number of large particle reduced when the ethanol was added to the fuel, this can be contributed to a slowdown of nuclear particles formation and growth.

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