Characteristic of Electron Density of Coaxial Dielectric Barrier Discharge of CH$_4$ /Air Mixture

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Abstract. Numerical study of dielectric barrier discharge of CH4/air mixture were carried out by employing a coaxial cylindrical electrode with diameter of 10mm under the condition of the discharge voltage of 6000V. The influence of electrode structure parameter and equivalence ratio on electron density were analyzed. The result shows that the electron density which increases with the decrease of the distance from the cathode has a quick increase at the small position near the cathode (0.05~0.4mm), and the maximum value of electron density appears at the position of 0.1mm. The electron density has no obvious change with the equivalence ratio, it is benefit to the lean combustion of natural gas application.

Introduction

As one of the main alternative energy sources of petroleum, the exploitation, storage and utilization of natural gas has aroused great interest. As the main component of natural gas, because of its tetrahedral molecular structure, methane has a high bond energy in C-H key [1,2], which leads to high ignition energy and low flame propagation rate [3]. Therefore, a high reaction temperature is necessary for the oxidation of methane. So the combustion reaction of methane is difficult to perform under low equivalence ratio conditions. In recent years, the non-equilibrium plasma enhanced combustion technology has received a wide range of attention since the ability to activate the molecules to cause a chemical reaction effectively.

Non equilibrium plasma ignition technology is the technology which ionizes the fuel or air into high energy electrons and heavy particles, thereby improving the reaction activity by applying an applied electric field. In the ionization mode of non equilibrium plasma, dielectric barrier discharge (DBD) is a kind of non equilibrium, unstable and non uniform gas discharge, which is a very effective method to generate non-equilibrium plasma [4,5]. However, only little literature could be found in the aspect of the effect of electrode structure parameter and equivalence ratio on the evolution law of electron of DBD. Therefore, numerical simulation was carried out to study the effect of discharge parameters and reactor structure on the distribution of electron density in the DBD discharge process of atmospheric pressure in this article.

Establishment of plasma model

The following assumptions have been made to establish the plasma kinetic model: 1) ignore the magnetic effect of the plasma; 2) Particle distribution in the ionization space is uniform; 3) The electric field distribution in the ionization space is uniform.

Physical model

Figure 1 is the schematic diagram of coaxial electrode structure, where the inner electrode is the anode and the radius is 3 mm. The outer electrode is the earth electrode. The space between the outer electrode and the inner electrode is ionization space, and its width S is 10mm. The dielectric layer thickness is 0.5 mm and the dielectric constant is 10. The initial electron density is 1.0×10^{14} m^{-3}, the initial electron energy is 4 eV, the secondary electron emission coefficient is 0.15, and the secondary electron energy is 5 eV.

Mathematical model

Arrhenius formula is as shown formula 1

$$k_f=AT^n\exp(-E_a/RT) \qquad (1)$$

The Surface reaction is shown as formula 2

$$A^++surface=>e+A \qquad (2)$$

The formula is shown as formula 3

$$-n\cdot\Gamma_e=(1-r)\cdot[(0.5v_{et}\cdot n_e)/(1+r)]-[\Sigma_i\,\gamma_i(\Gamma_i\cdot n)+\Gamma_t\cdot n] \qquad (3)$$

where n represents the total particle number density; n_e represents the number density of electrons; Γ_e represents the diffusion term; r indicates the reflectivity of the particles bombarding; v_{et} is the velocity of electron thermal motion; Γ_i stands for the ith positive ion flux at the wall; γ_i represents the secondary electron emission coefficient of wall reaction of the ith positive ions; Γ_t is the gamma emission flux of wall heat.

In the computational model, the mixture of the gas composition is mainly considered as CH_4, N_2 and O_2 that carried out at atmospheric pressure and initial temperature of 298K.The reactions are shown in Table 1 and Table 2.For the surface reaction are as shown in table 3, heavy particles could excite secondary electron by wall-colliding. According to the physical model of figure 1, a one-dimensional plasma model as shown in figure 2 has been established. In the model, the horizontal axis represents the distance L from the center electrode to the outer electrode along the radius direction, and the unit is mm.
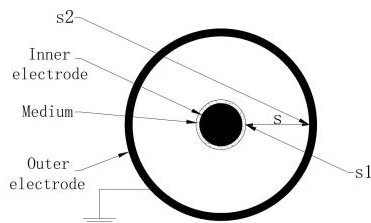

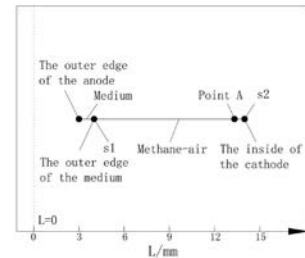Fig. 1 Schematic diagram of coaxial cylinder electrode structure   Fig. 2 One dimensional plasma model

Table 1 Chemical reactions of non electronic components

Reaction	Reaction equation	A	n	En
1	$N+O_2\rightarrow NO+O$	9×10^9	1	6500
2	$N+NO\rightarrow N_2+O$	2.7×10^{13}	0	355
3	$CH_4+O\rightarrow CH_3+OH$	4.7×10^{-10}	0	5.4×10^4
4	$CH_2O+O\rightarrow HCO+OH$	3×10^{-11}	0	1.2×10^4
5	$H+O_2\rightarrow OH+O$	1.6×10^{-10}	0	6.4×10^4
6	$CH_4+1.5O_2\rightarrow CO+2H_2O$	5×10^{11}	0	2×10^5
7	$CO+O\rightarrow CO_2$	2.24×10^{19}	0	1.7×10^5
8	$O_3+N_2\rightarrow O_2+O+N_2$	4×10^{14}	0	22667
9	$O_2+O+N_2\rightarrow O_3+N_2$	1.6×10^{14}	-0.4	-1391
10	$O_3+O_2\rightarrow O_2+O+O_2$	1.54×10^{14}	0	23064

Table 2 Electron impact reaction in methane air reaction

Reaction	Reaction equation	Reaction rate	$\Delta\varepsilon$/eV
1	$e+O_2 \rightarrow e+O_2(ald)$	Collision cross section calculation	4.5
2	$e+O_2 \rightarrow 2e+O_2^+$	Collision cross section calculation	12.06
3	$e+N_2 \rightarrow e+N+N$	Collision cross section calculation	13
4	$e+CH_4 \rightarrow e+CH_3+H$	Collision cross section calculation	10

Table 3 surface reaction

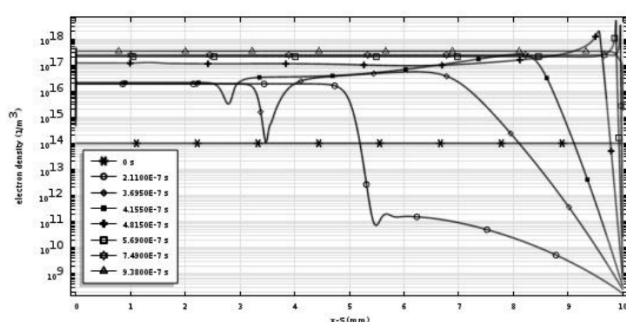| Reaction	Equation	Adhesion coefficient
1	$O_2^+ \rightarrow O_2$	1
2	$O \rightarrow 0.5O_2$	1
3	$N_2^+ \rightarrow N_2$	1

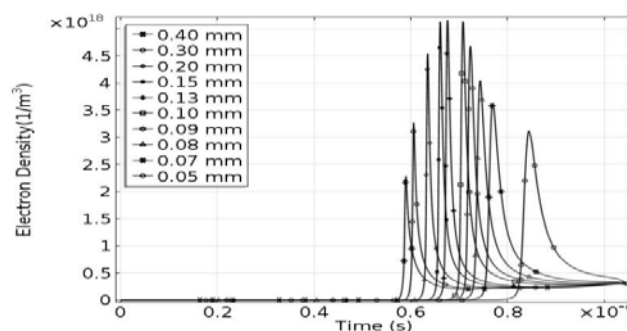Fig.3 The space distribution of electron density

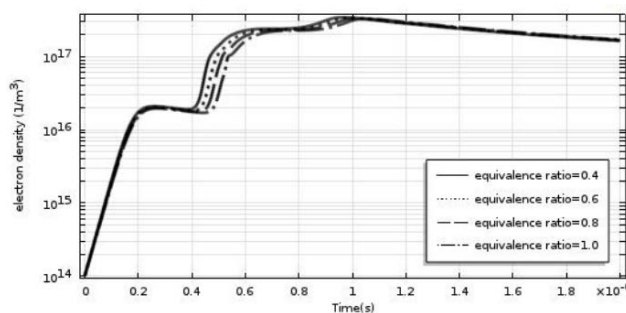Fig.4 The electron density at different distances from the cathode

Fig.5 The electron density under different equivalence ratios at a distance of 0.12 mm from the anode
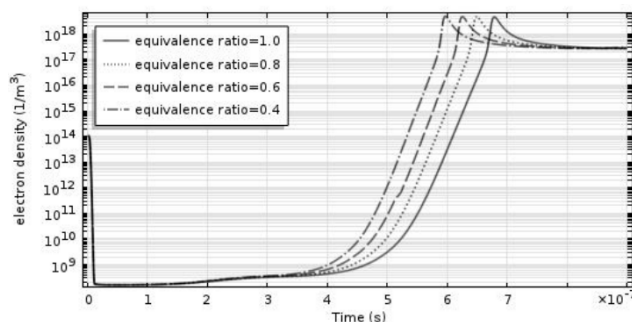

Fig.6 The change of electron density with time at different equivalence ratio conditions

Results and discussion

Figure 3 shows the distribution of electron density in the ionization space, in which each curve represents the distribution of electron density at a time in the discharge gap. In this research, the voltage applied to the internal electrode is 6000 V, and the discharge gap is 10 mm.

It shows that the electrons accumulate to the anode rapidly at first, and then there is an sharply increase in electron density around cathode. This is because the free electron are accelerated when moving from the cathode to the anode under applied electric field since discharge. During the acceleration, most of the electrons and neutral molecules occur elastic collisions, but a small part of high energy electrons and neutral particles occur inelastic collisions. This leads to the collisional ionization and large number of electron avalanche. Therefore, the number of electrons grows exponentially. In addition, the bombardment by heavy particles (mainly positive ion) to the cathode could lead to the secondary electron emission at cathode surface. It will produce more electrons,

which leads to the drastic change of electron density near the cathode. As a result, the points with the distance from the cathode as 0.4 mm ~ 0.05 mm have been picked as the research point.

The variation of the electron density with time is shown as Fig.4. It shows that the electron density appears a trend of rapid increase followed by a sharp decline. This is due to the bombardment of the heavy particles on the cathode has initiated secondary electron emission, which leads to an increase in the density of electrons in the vicinity. However, with the decrease of distance from the cathode (between 0.4 mm and 0.05 mm), the maximum value of the electron density firstly increases and then decreases. This is caused by many factors: on the one hand, when the distance is very close to the cathode (less than 0.05 mm), the electron that has just been bombarded has high energy and velocity, it keeps colliding with the heavy particle and leads to the velocity reduction of a part of electrons; on the other hand, the collision between electron and positive ion, as well as ground state particle and excited particle have produced new electrons; and also, some of the low-energy electrons will be adsorbed or neutralized by heavy particles. The combined effect of these factors makes the electron density reaches to the maximum value at a distance of 0.12 mm from the cathode.

Figure 5 shows the variation of the electron density with time at a distance of 0.12 mm from the anode. It shows that the change of electron density with time appears three stage under different equivalence ratio conditions: the first stage is 0~0.5 μs, where the electron density significantly increases and gradually decreases at 0.25 μs; the second stage is 0.5 ~ 1.0 μs, where the electron density increases significantly at first, then the increment speed gets slower and reaches the maximum at 1.0 μs; the third stage is 1.0 ~ 2.0 μs, where the electron density decreases gradually. The change of electron density in the first stage is caused by the initial electron in the ionization space, while the change in the second stage is mainly due to the secondary electron. Then the electron density starts to decrease after reaching to the maximum value since the effect of the internal electric field.

Figure 6 shows the effect of equivalence ratio on the change of electron density. The variation trend of electron density is basically the same under different Φ conditions, only that the electron density could reach the maximum value faster at low Φ. It could also be obtained that although the equivalence ratio has been reduced, it has no obvious influence on the electron density which plays a key role in active particle production. It has a very beneficial effect on the methane-air combustion reaction under the conditions of low equivalence ratio.

Conclusion

(1) In the ionization space, the electron density increases with the decrease in the distance from the cathode, and the maximum value of the electron density increases at first and then decreases, and reaches to the maximum value at a distance of 0.12 mm.

(2) There is no obvious change in the electron density with the equivalence ratio, which has a very beneficial effect on the methane-air combustion reaction under the conditions of low equivalence ratio.

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