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Theoretical analysis on reaction process of active species in atmospheric pressure streamer discharge

シミュレーションによる大気圧ストリーマ放電の活性種生成機構解明

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A practical streamer discharge model is developed to analyze the characteristic of pulsed streamer discharge in a positive point-to-plane electrodes filled with air at room temperature and atmospheric pressure. The calculated primary and secondary streamer development shows good agreement with an experimentally obtained streak photograph. The experimentally obtained axial distribution of oxygen and nitrogen radical productions by streamer discharge can be successfully reproduced in our simulation

1. Introduction

Streamer like discharges is widely used in many applications such as decomposition of gaseous pollutant, water treatment, ozone production, surface treatment and medical application. However, the understanding of the phenomena is still poor. The complete simulation of the streamer discharge can lead to a better understanding of the physico-chemical activity and help us to choose the best operating conditions (such as reactor geometry, the flue gas resident time, the applied voltage shape and magnitude) in order to improve the process efficiency. To perform quantitative analysis of the efficiency of various applications, an insight is needed into the physics of streamer propagation processes. For one of the important factors in streamer discharge phenomenon, it is known that the vibrationally excited molecules have large influence on the discharge phenomenon.

2. Kinetic model

The propagation of a streamer discharge is considered in a point to plane electrode configuration with a gap distance of 13mm. In order to simulate the discharge development, we chose a cylinder domain of 18cm height and 8.0cm radius. The total number of grid is Nz×Nr = 1972×256 with spatial steps from dz = 1 μm (near point and plane) to 10 μm (inter-electrode gap) for z-axis and from dr = 2.5 μm up to 200 μm for r-axis. The point electrode has a revolutionary hyperboloid shape with a radius of 40μm. The simulation conditions are summarized in Table.1.

To compute the streamer propagation in air, we use first-order electro-hydrodynamic model for electrons, positive and negative ions in the framework of the drift-diffusion approximation.

Table.1. Simulation condition

Temperature	300 K
Pressure	101 kPa
Gas components	Dry Air
Gap length	13 mm
Point curvature radius	40 mm
Time step	2.5×10^{-13} s
Initial electron density	10^{3}cm^{-3}

Thus, the equations involved in this model are following

$$\frac{\partial}{\partial t}n(r,t)+\nabla_r[n(r,t)v(r,t)]=S(r,t)\ldots(1)$$

$$\frac{\partial^2\phi}{\partial r^2}+\frac{1}{r}\frac{\partial\phi}{\partial r}+\frac{\partial^2\phi}{\partial z^2}=\frac{q}{\varepsilon_0}(n_p-n_e-n_n)\ldots(2)$$

where, *n(r,t)* is the charged particle density, *S(r,t)* is the particle chemical source term, *v(r,t)* is the particle velocity, ϕ is electric potential, ε_0 is the permittivity of free space, q is the absolute value of the electronic charge. The subscripts of *e*, *p* and *n* stand for electron, positive and negative ions, respectively. The transport and source parameters (such as particle mobilities and reaction coefficients) are calculated using Bolsig+ [1] with published e-V cross sections [2].

Photoionization is taken into account through the three-exponential Helmholtz models derived by Luque *et al*. [3]. The charged species transport equations are solved with the MUSCL superbee algorithm. To solve the Poisson's equations and photoionization term, we use Red&Black Multigrid method and calculate it on GPU (Graphics Processing Unit). Due to this powerful GPU computing method, the performance of solving

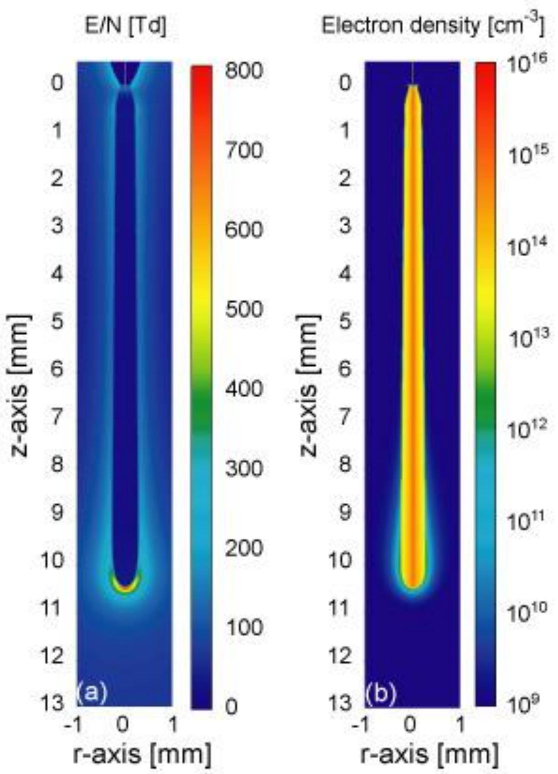

Figure.1. Two dimensional (a) reduced electric field *E/N* (in Td) and (b) electron density (in cm^{-3}) for 13 mm gap discharge at time t = 20 ns.

Poisson's equation achieves 40 times speed up compared with 1 core CPU calculation.

3. Results

Figures.1 shows the 2-D structure of the absolute value of the reduced electric field and the electron density for 13 mm gap point to plane discharge. Figure.1(a) indicates that the streamer head propagates with a high electric field toward the cathode and streamer channel corresponds to a highly conductive channel connecting the streamer head to the anode. The reduced electric field is equal to 20 Td in the streamer channel and around 800 Td in the streamer head. Figure.1(b) shows that the electron density in the streamer channel is higher than $10^{14}cm^{-3}$. From both figures, the streamer channel radius, which is defined as the radial extension of low reduced electric field area in the streamer channel, is estimated at 300 μm and the mean streamer velocity is around 4.3×10^5 m/s. These results show good agreement with our experimental results [4].

Figures.2 show the axial distribution of O radical densities at t = 100 ns and streamer luminous intensities for V = 32, 24 and 16 kV. In figures.2, the axial distribution of oxygen radical densities and streamer luminous intensities are roughly in good agreement. This characteristic has been

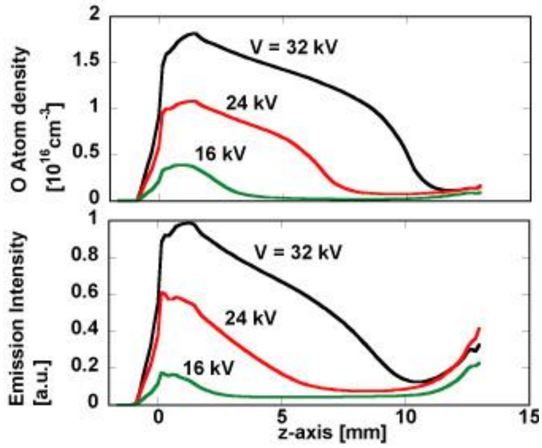

Figure.2. Axial distribution of oxygen radical density at t = 100 ns and streamer luminous intensity for V = 32, 24 and 16 kV.

already obtained from out experimental results [5].

For N radical production, it is generally considered that N radicals are produced by following reactions [6]

$$N_2(X^1\Sigma_g^+) + e \rightarrow N_2^* + e$$
$$\rightarrow N(S) + N(D) + e$$
$$N_2^* = (\text{sum of thesingret states})$$

However, these reactions alone cannot explain the relatively high dissociation degrees observed experimentally [7]. Therefore, many other processes for nitrogen dissociation have been suggested. The following mechanism is known as one of these processes [8]

$$N_2(X^1\Sigma_g^+, v) + e \rightarrow N_2(B^3\Pi_g^+, v'>12) + e$$
$$N_2(a^1\Pi_g^+, v'>6) + e$$
$$\rightarrow N(S) + N(S, D) + e$$

These reactions closely related to the vibrationally excited molecules of nitrogen and are efficient in low energy region. We can successfully reproduce our experimental data by these processes. In atmospheric pressure streamer discharge, vibrationally excited molecules have great influence on the production of N radicals.

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