Numerical study on propagation mechanism and bio-medicine applications of plasma jet

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Abstract: In this study, the propagation mechanism of plasma jet and some bio-medical applications are investigated by two-dimensional numerical model. The key equations of plasma physics and chemistry related with plasma jet are firstly introduced. The simulation results suggest that the sheath forms near the dielectric tube inner surface, which results in the plasma channel to shrink in the radial direction inside the dielectric tube. The photoionisation of air species plays a crucial role in the transition from the localised discharge to streamer. The Penning ionisation increases the electric conductivity of the plasma channel and facilitates the formation of ring-shaped plasma bullet. For the plasma jet in the open air, electron-impact dissociation of H2O, electron neutralisation of H2O+, as well as dissociation of H2O by O(1D) are found to be the main reactions to produce OH. For micro plasma jet, the higher ignition voltage as the tube diameter decreased is attributed to the deceasing pre-avalanche electron density inside the tube. The simulation of plasma treatment of bacteria biofilm indicates that the mean free path of charged species in µm scale permitted the plasma penetrate into the cavity of the biofilm, and the structure of the biofilm results in the non-uniform distribution of ROS and RNS. The simulation of plasma jet treatment of cells immersed in liquid suggests that the HO2 generated by plasma aqueous species is the only way for superoxide to penetrate cell membrane and damage cytosolic fumarase B.

1 Introduction

Atmospheric-pressure plasma jets, formed by gas discharges in noble gas flow inside a dielectric tube and injected into ambient air, have shown significant promising applications such as biomedicine and nano-materials processing. The plasma jets usually consist of a streamer head travelling at a speed of 105–106 cm/s. During the streamer head advancement, chemical reactions take place in the fluid mechanical mixture layer where noble gas and air inter-diffuse, so numerous reactive species such as reactive oxygen species (ROS) and reactive nitrogen species (RNS) form and are delivered 1–20 cm downstream. Recently, many experimental diagnostic methods have been published to understand the propagation mechanism of plasma jet. Ring-shaped profiles of streamer head radiation were revealed by nano-second exposure time imaging, and the maximum concentration of ions and reactive species are shifted from the plasma jet axis [1–4]. The experiments show that the radial distribution of the axial electric field is also ring-shaped [5]. Simultaneously, to understand these experimental observations, many computational models were also developed. By assuming different initial electron density profiles, Naidis developed a two-dimensional (2D) model and realised the transition of the streamer head from disk to ring shape [6]. Sakiyama et al. [7] got the helium-nitrogen distribution along the jet axis by using the neutral gas flow equations, and they found that Penning ionisation plays a critical role in the formation of ring-shaped luminous region. The properties of the helium streamer and of the plasma channel behind the streamer head as a function of parameters such as electrode geometry, voltage pulse waveform and pre-ionisation density are discussed by Boeuf et al. [8]. Liu et al. found the impurities of working gas through photoionisation have significant effects on the discharge propagation speed inner tube [9]. However, these experimental and numerical results neglected the discharge dynamics within the dielectric tube which is crucial for the propagation of plasma jet.

On the other hand, understanding the production mechanism of reactive species in the plasma jet is also quite important to improve treatment efficiency of the plasma jet [10–13]. By using cavity ring-down spectroscopy and laser-induced fluorescence (LIF), the absolute density of OH species is measured. The OH density ranged from 1.3 × 1012 to 4.6 × 1016 cm−3 in the Ar, Ar/N2 and Ar/O2 atmospheric-pressure plasma jets, is measured by Wang and Srivastava [14]. Through the transition line P1(4) from OH(N, v′ = 1, j′ = 3) → OH(X, v′ = 0, j′ = 4) which used for the LIF excitation, the absolute OH density was determined to be 2.5 × 1016 m−3 at 1 mm away from the jet nozzle [15]. The concentration of OH radicals on atmospheric-radiofrequency plasma jet calibrated by Rayleigh scattering on ambient air was measured by Vorač et al. [16] and the maximum OH density near the tip of the visible active dischage was found approximately to be 5 × 1020 m−3. Spatially resolved OH LIF signals show that the signal intensity is stronger on both edges, which gives rise to the donut shape of the OH distribution [17]. The simulation results suggest that the main reactions of OH in an atmospheric-pressure streamer discharge are the dissociation reactions of H2O with O (1D) and N (a′′ → X′′) [18]. The modelling of the He–H2O plasma jet driven by pulsed DC high voltage revealed that the dominant source of OH radicals is related to the Penning and charge transfer reactions of H2O molecules with excited and charged helium species [19]. More numerical models with matching of experimental measurement are still needed to understand the production mechanism of reactive species.

Recent developments in plasma source have shown significant promising applications such as food decontamination and cancer cell treatment. These plasma sources are very effective in treating a very small target such as the single cancer cell treatment [20] and endoscopic surgeries [21, 22], or generating a wide range of chemically active species (ROS and RNS) which used for food decontamination and blood coagulation [23–34]. Niemira and Sites [23] used cold plasma and successfully inactivated Salmonella
Stanley and Escherichia coli O157 on apples. Pathogenic organisms and spoilage organisms on the pericarpis of mangoes and melons are also inactivated by cold atmospheric plasmas [24]. Plasma is also applicable in surface decontamination of pre-packed RTE meat product (bresaola) inoculated with Listeria innocua [25]. More simulation work is needed to understand physics and chemistry related with plasma treatment.

For the living tissues and cells are immersed in liquids (such as blood or culture media), the interactions of plasma with liquids are a complicated process [35]. Generally the reactive species generated by plasma are dissolved to be different types of ROS/RNS in liquids. Hamaguchi [27] developed a numerical code to study the time evolution of various charged and charge-neutral species generated by low temperature plasma, including hydrogenated electrons, in pure water. Chen [28] presented the development of a model framework for plasma-biofilm and plasma-tissue interactions that can link molecular simulation of plasma chemistry to function at a cell population level or a tissue level. Tian and Kushner [29] reported on a computational investigation of the interaction of dielectric barrier discharges (DBDs) in humid air with a thin water layer covering tissue, and the simulation results suggest that the photolysis of water by plasma-produced UV/VUV plays a significant role in the production of radicals, and the dominant reactive oxygen and nitrogen species (RONS) in the liquid are O₃,aq, H₂O₂,aq and HO₂,aq (aq means aqueous species). Bruggeman and co-workers investigated the plasma-induced liquid chemistry by performing liquid ion chromatography and hydrogen peroxide concentration measurements on treated distilled water samples [30]. Ikawa et al. [36] and Pei et al. [37] show that the process of plasma treatment is in minutes scale and the eventual intracellular concentration of reactive species related to plasma is difficult to measure. Therefore, the delivery and transform the process of plasma active species into cells immersed in liquids also need be investigated.

In this paper, the propagation mechanism of plasma and some plasma applications models will be studied. The paper is organised as follows: First, the numerical model is presented, including plasma governing equations, three-term exponential Helmholz photo-ionisation model, plasma chemistry, and the Navier–Stokes, continuity and convection–diffusion equations in order to describe the gas flow from the nozzle. The computational model of the interaction between the plasma and cells immersed in the culture media is also introduced. Second, the plasma jet propagation mechanism and some plasma applications models are discussed. The main mechanism of streamer propagation is investigated, and as the tube diameter changed, the influence for the transition mechanism is also studied. Finally, some latest simulation advances such as plasma treatment of bacteria biofilm, plasma treatment of cells immersed in liquid are presented.

2 Numerical model

2.1 Plasma governing equations

The plasma governing equations are consists of a series of coupled equations for the electric field and for the densities of charged and uncharged species [27].

2.1.1 Species continuity equations: The number densities of the species are obtained by solving separate species continuity equations using the drift–diffusion approximation

\[
\frac{\partial n_s}{\partial t} + \nabla \cdot \Gamma_i = R_i, \quad s = 1, 2, \ldots, k_g
\]

where \(n_s\) is the species density, \(\Gamma_i\) is the species number flux, \(R_i\) is the individual species chemistry source, which are computed using a mass-action kinetics formulation with reaction pathways and reaction coefficients. The index \(s\) represents all the species considered.

The flux of the different species is expressed as follows

\[
\Gamma_i = -\mu_i \nabla \Phi - D_i \nabla n_s
\]

Here, \(\mu\) is the mobility and \(D\) is the diffusion coefficient for the corresponding species, for the neutral species, the flux is only determined by diffusion term. \(\Phi\) is the electric potential.

2.1.2 Poisson equation: The electric field \(E\) is governed by the Poisson equation for the potential \(\Phi\)

\[
E = -\nabla \Phi
\]

\[
\nabla \Phi = -\frac{\rho}{\epsilon_0}
\]

where \(\rho = e (n_i - n_e)\) is the volume charge density, \(n_i\) represents the number density of positive ions and \(n_e\) is the number density of electrons.

2.1.3 Electron energy conservation equation: The electron temperature in the discharge is calculated using the electron energy conservation equation

\[
\frac{\partial \rho_e}{\partial t} + \nabla \cdot \rho_e (\vec{v}_e \rho_e) = \frac{3}{2} k_b T_e \frac{2 m_e}{n_e} (T_e - T_g) \varepsilon_e \chi_{eb}
\]

The left-hand side of the electron energy equation consists of two terms: the mean electron energy (averaged over the energy distribution function), energy mobility and diffusivity. The right-hand side consists of three source terms: Joule heating, inelastic collisional heating and elastic collisional heating. \(\varepsilon_e\) is the mean electron energy, \(\varepsilon\) is the electron energy mobility, \(\Gamma_i\) is the electron energy flux, \(D_e\) is the electron energy diffusion coefficient, \(\epsilon\) is the unit electron charge, \(\Delta E^e\) is the energy loss in the reaction i, \(\gamma\) represent the reaction rate of reaction i, \(m_i\) is the mass of mass and \(\chi_{eb}\) is the species momentum transfer collision frequency.

2.2 Photoionisation model

The photoionisation model is based on the three-term exponential Helmholz model developed by Bourdon et al. [38]. The three-term exponential Helmholz model is an approximation of classical integral model derived by Zhelezniak et al. [39]. According to the classical integral model, for photoionisation in air, the photoionisation rate of oxygen molecules (\(O_2 + hv \rightarrow e + O_2^+\)) at point of observation \(r\) due to the emission of ionising radiation at all source points \(r\) is given by an integral equation

\[
S_{ph}(r) = \int \int \int \frac{I(r')g(R)}{4\pi R^2} dV'
\]

where \(R = |r - r'|\), \(I(r')\) is the intensity of radiation from emitting species (\(e + N_2 \rightarrow e + N_2^+ + hv\) in this case). \(I(r')\) is proportional to the ionisation rate \(\Sigma_i(r')\) of the emitting species multiplied by a factor \(\xi\) (\(\xi = 0.02\)) and a quenching factor \(P_q/I_{ph}\), where \(P_q\) is (\(30\sim 60\) Torr) and \(P\) is the gas mixture pressure [40]. \(g(R)\) is absorption function, it can be fitted from the experimental data using (8)

\[
I(r') = \frac{P_q}{P + P_q} \xi S_i(r')
\]

\[
g(R) = \exp \left[ -\frac{\lambda_{max} \rho_e R}{P_q} \right] - \exp \left[ -\frac{\lambda_{min} \rho_e R}{P_q} \right]
\]

where \(\lambda_{max}\) and \(\lambda_{min}\) are fitting parameters.

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Bourdon proposed a new method to reduce the computational cost of the integral model. It is called three-term exponential Helmholtz model. ‘Three-term’ for it divides the source ionisation into three terms with relevant fitting data and parameters for air [38]. Therefore the form is

\[ S_{\text{ph}}(r) = \sum_{j=1}^{3} S_{\text{ph}}^j(r) \]  

with each photoionisation source term have the form

\[ S_{\text{ph}}^j(r) = \int \int \int \frac{j(r')}{4\pi r'^2} A_j P_{O_2} c^{-j} P_{O_2} R \]  

satisfying the Helmholtz differential equations

\[ \nabla^2 S_{\text{ph}}^j(r) - (\lambda_j P_{O_2})^2 g S_{\text{ph}}^j(r) = -A_j P_{O_2}^2 f(r) \]  

Thus it can be easily derived by comparing (6) and (10)

\[ g(r) = P_{O_2}^2 R \sum_{j=1}^{3} A_j c^{-j} P_{O_2} R \]  

with the photoionisation parameters \( A_j \) and \( \lambda_j \) [38].

### 2.3 Plasma chemistry

Plasma chemistry includes helium chemistry, helium–air chemistry and air chemistry reaction sub-mechanisms when working gas is helium gas. It can be argon gas, air and others depending on the working gas. All these reactions are taken from the literature [41–43]. For reactions which produced \( N_2 \) and \( O_2 \) in different rotational and vibrational states, their reaction rates are calculated with the photoionisation parameters \( A_j \) and \( \lambda_j \) [38].

### 2.4 Gas flow equations

For a proper description of the plasma jet, the gas flow is described. The gas flow along the dielectric tube is calculated by Navier–Stokes, continuity and convection–diffusion equations. Therefore, the influence of gas flow to the plasma displacement can be quantised

\[ \rho_g \frac{\partial u_g}{\partial t} + \nabla \left[ p_g I - \mu_g (\nabla u_g + (\nabla u_g)') \right] = F \]  

\[ \rho_g \nabla u_g = 0 \]  

\[ D \nabla^2 c - u_g \nabla c = 0 \]  

where \( \rho_g \) is the gas density, \( p_g \) is the gas pressure, \( \mu_g \) is gas viscosity (helium: \( 1.94 \times 10^{-5} \) Pas and air: \(1.94 \times 10^{-5} \) Pas), \( I \) stands for the unit matrix and the superscript ‘T’ represents the tensor transpose operation, \( D \) is the diffusion coefficient (helium in air \( 7.2 \times 10^{-7} \) m²s⁻¹), \( c \) is the helium mole fraction mol·m⁻³ and \( F \) is the body force (buoyancy) [45].

However, the gas flow equations are not solved together with plasma governing equations always because of the limits of computer memory. Instead, one method is they are first solved separately, and then, the obtained velocity distribution is used as input data [46]. The other method is we can get a stationary laminar velocity field after enough time because of small simulation space outside the tube that the carrier gas, helium gas, e.g. output from the nozzle

\[ \rho_g \frac{\partial u_g}{\partial t} = 0 \]  

Compared with formula (13),

\[ \nabla \left[ p_g I - \mu_g (\nabla u_g + (\nabla u_g)') \right] = F \]  

using this equation to calculate the final velocity distribution as input data.

### 2.5 Mass balance in the liquid

Since the ‘plasma medicine’ such as the plasma treatment of liquid has been widely used, in order to describe the transport of plasma reactive species into the liquid, the mass balance model in the liquid developed by Maier et al. [47] is introduced. In this model, the concentration profile \( C_{\text{plasma,aq}}(x, t) \) (’aq’ means aqueous species) is

\[ \frac{\partial C_{\text{plasma,aq}}}{\partial t} = D_c \frac{\partial^2 C_{\text{plasma,aq}}}{\partial x^2} - k_1 C_{\text{plasma,aq}} C_1 \]

\[ - k_2 C_{\text{plasma,aq}} C_2 - \cdots \]

with the boundary conditions (see Fig. 5)

\[ \text{BC1: } C_{\text{plasma,aq}}(x = 0) = C_{\text{plasma}}^a \]

\[ \text{BC2: } \left[ \frac{\partial C_{\text{plasma,aq}}}{\partial x} \right]_{x=L} = 0 \]

where \( D_c \) is the diffusion coefficient in liquid and \( k_i \) is the rate coefficient of water solvation reactions. The reaction between aqueous plasma species and cells at the moment is not included for the concentration of cells in the culture media and the reaction rate between aqueous plasma species and cell as a unit is unknown.

The concentration profile \( C_{\text{plasma,aq}}(x, t) \) which depends on the plasma contact time and the penetration depth is shown in Fig. 1. The boundary condition \( C_{\text{plasma}} \) at the gas–liquid interface can be

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**Fig. 1** Dissolved plasma concentration profile in the liquid. \( C^* \) plasma is the concentration of plasma reactive species at gas–liquid interface. \( L \) is the thickness

**Fig. 2** Schematic of model system predicting intracellular concentration of plasma reactive species

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calculated by

\[ C_{\text{plasma}} = k_H \left[ C_{\text{plasma, gas}} - \frac{1}{D} \cdot \Gamma_{\text{plasma, gas}} \cdot \Delta t \right] \]  \hspace{1cm} (21)

where \( k_H \) is Henry’s constant, \( D \) is the diffusion coefficient of plasma reactive species, \( \Gamma_{\text{plasma, gas}} \) is a concentration flux of neutral plasma species. It can be estimated by the simulation model \([45]\). The equivalent density at the gas–liquid boundary is calculated according to the condition of \( C_{\text{plasma}} \leq C_{\text{gas}} \cdot k_H \) \([29]\). According to Henry’s law \([27]\), it can be deduced that the dissolved plasma species is proportional to its partial pressure in the gas phase, thus the outward flux from water to the gas phase is obtained.

2.6 Species continuity equation in cells

Getting the concentration and distribution of the dissolved plasma active species from liquid into the target cells is the important one of the purposes we model the plasma treatment of liquid. However, target cells always have a variety of shapes. The idealised method is to assume the target cell is surrounded by a stagnant film of thickness \( \delta (\leq a) \) with a steady, spherically symmetric concentration field (Fig. 2b) \([48]\). Thus, for the active species \( i \), the conservation equation for species \( i \) at radial position \( r \) in the cytosol is

\[ -\frac{D_i}{r^2} \frac{dC_i}{dr} + m_i + (1 - \phi)R_i = 0 \]  \hspace{1cm} (22)

with the boundary condition

BC1 (at the cell centre symmetry boundary):

\[ \frac{dC_i}{dr} \bigg|_{r=0} = 0 \]  \hspace{1cm} (24)

BC2 (at the cell membrane):

\[ -\frac{D_i}{r^2} \frac{dC_i}{dr} \bigg|_{r=a} = \frac{P_i}{\rho R_i} \left[ C_i(a) - k_i C_i(a) \right] = -D_i \frac{dC_i}{dr} \bigg|_{r=a} \]  \hspace{1cm} (24)
where $D_i$ is the effective cytosolic diffusivity, $C_i$ is the concentration of species $i$ in the cytosol, $m_i$ is the rate of entry by diffusion from the mitochondria, per unit cell volume, $\phi$ is the mitochondrial volume fraction and $\bar{R}_i$ is the rate of formation by chemical reaction. The differential equations for the mitochondria and the liquid film are analogous to (22), but without $m_i$ or the factor $1 - f$. Equation (24) is a result of equality of fluxes at the cell membrane, where $\bar{P}_i$ is the cell membrane permeability, $\bar{k}_i$ is the ratio of intracellular to extracellular concentrations at equilibrium.

### Table 1 Chemistry reactions of H$_2$O

<table>
<thead>
<tr>
<th>Reaction no.</th>
<th>Reaction</th>
<th>Reaction rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>R1</td>
<td>$e + H_2O \rightarrow 2e + H_2O^+$</td>
<td>Bolsig+</td>
</tr>
<tr>
<td>R2</td>
<td>$e + H_2O \rightarrow e + H + OH$</td>
<td>Polsig+</td>
</tr>
<tr>
<td>R3</td>
<td>$e + H_2O^+ \rightarrow H + OH$</td>
<td>$7.11 \times 10^{-4}$ $T^{-0.5b}$</td>
</tr>
<tr>
<td>R4</td>
<td>$e + H_2O^+ \rightarrow O + 2H$</td>
<td>$3.1 \times 10^{-3}$ $T^{-0.3b}$</td>
</tr>
<tr>
<td>R5</td>
<td>$H + OH + M^* \rightarrow 2OH$</td>
<td>$4.3 \times 10^{-25}$</td>
</tr>
<tr>
<td>R6</td>
<td>$O_{tot} + H_2O \rightarrow 2OH$</td>
<td>$2.2 \times 10^{-10}$</td>
</tr>
<tr>
<td>R7</td>
<td>$2OH \rightarrow O + H_2O$</td>
<td>$2 \times 10^{-12}$</td>
</tr>
<tr>
<td>R8</td>
<td>$O + OH \rightarrow H + O_2$</td>
<td>$3.32 \times 10^{-11}$</td>
</tr>
<tr>
<td>R9</td>
<td>$O + H + M^* \rightarrow O + H + M^*$</td>
<td>$1.62 \times 10^{-33}$</td>
</tr>
<tr>
<td>R10</td>
<td>$H + O_2 \rightarrow OH + O$</td>
<td>$3.55 \times 10^{-22}$</td>
</tr>
<tr>
<td>R11</td>
<td>$OH + M^* \rightarrow OH + M^*$</td>
<td>$4 \times 10^{-3} \exp(-415700/RT)^b$</td>
</tr>
<tr>
<td>R12</td>
<td>$O + H_2O \rightarrow 2OH$</td>
<td>$2.34 \times 10^{-15}$</td>
</tr>
</tbody>
</table>

Units: Two-body reaction rate coefficient (cm$^3$ s$^{-1}$) and three-body reaction rate coefficient (cm$^6$ s$^{-1}$)

$^a$Species $M$ in reactions R5, R9, R11 represents third-body species

$^b$The tabulated reaction rates are given in the Arrhenius form

Fig. 6 Propagation dynamics of the streamer

a Ionisation rate of plasma jet at 50 ns
b 5 ns exposure time image of plasma jet at 100 ns
c Ionisation rate of plasma jet at 70 ns (simulation)
d 5 ns exposure time image of plasma jet at 150 ns
e Enlarged image of ionisation inside the tube [red dashed circle in (a)]
f Enlarged image of ionisation inside the tube [red dashed circle in (d)]

3 Recent advances in plasma simulation

3.1 Plasma jet propagation dynamics

Fig. 3a shows the structure of 2D plasma jet simulation domain. The simulation domain is 10 × 20 mm. The radius of dielectric tube is 5 mm and the inside diameter of tube is 2 mm. The tube embeds a single cylindrical power electrode, and the length of the electrode is 1.5 mm, the thickness of electrode is 0.2 mm. The amplitude of pulsed dc voltage applied in the electrode is 2000 V and the rising time is 50 ns.

The helium mole fraction is shown in Fig. 3b. The gas flow rate is 1100 standard cubic centimetres per minute (SCCM). Diffusion and convection of gas cause the appearance of ‘mixing layer’, where the

Fig. 7 Distribution of electron density, electron energy and electric field

a Electron density at 120 ns
b Electron energy at 120 ns
c Electric field of the plasma jet at 120 ns
d Electron density at 180 ns
e Electron energy at 180 ns
f Electric field of the plasma jet at 180 ns
He–air chemical reactions occur. The helium mole fraction gradually decreases in the vertical direction away from the tube axis, and in turn, the proportion of air increases (Fig. 3c).

The electron density distribution shown in Fig. 4b has the same profile with the photo shown in Fig. 4a. The self-consistent, multi-species, 2D axially symmetric plasma model successfully explains the plasma inside the tube has a necked profile in the radial direction, while the plasma outside the tube has a ring-shaped streamer head with the weaker optical emission along the tube axis [49].

The number density of electrons and He positive ions are shown in Fig. 5d. The positive ion density is six orders of magnitude higher than the electron density suggests the existence of the sheath when the radial position is from 4.7 to 4 mm. As a result of the lower mobility of ions than electrons, a strong positive electric field from the inner surface of tube to the boundary of plasma channel forms (Fig. 5c), and it results in electron repulsion which helps to facilitate the positive space-charged region (sheath), and the neutral plasma is shielded from the part of the tube’s inner surface [50–52]. A large number of seed electrons are produced by metastable reactions and the impact of ions on the tube’s inner surface, and the sheath provides high energy for these electrons as a result of electrons are accelerated in the strong electric field. Therefore, the ionisation in the sheath is much lower than the ionisation occurs in a plasma channel (Figs. 6a–f) [53]. When the streamer grows to the position of the tube nozzle, the sheath retreats and the plasma expands in the radial direction. A possible explanation why sheath decreased is that the streamer head is far away from the electrode. Fig. 6 shows the off axis distribution of peak ionisation region when the streamer exits the nozzle, and it reveals the ring-shape plasma bullet.

After the streamer exceeds the tube nozzle, the avalanche-to-streamer transition mechanism guides the subsequent advancement of streamer. According to the Meek’s criterion [54, 55], the streamer moved forward because of the electric field pushed by charge in ionisation region retreated in the direction forward to the ground.

In the He–air mixing layer, the consumption of energy from power electrode coupled to the streamer head by reactions causes the shrink of plasma channel in the radial direction (Figs. 7a, c). The simulation also suggests the electron impact ionisation has smaller contribution to the number density of the positive ions compared to the Penning ionisation. Therefore, the electric conductivity of the plasma channel is enhanced by Penning ionisation. It also helped the ring-shaped streamer propagation because of severe Penning ionisation in the mixing area.

Due to the OH is one of the most active species generated in moist gas mixtures, the production mechanism of OH radicals in a pulsed DC plasma jet discharge is studied numerically. The simulation results suggest that the electron-impact dissociation of H2O is the most important reaction to produce the OH species. The
recombination of H$_2$O$^+$ by electrons and the dissociation of H$_2$O by O(1D) are also contributing much to produce OH radicals. On account of Penning ionisation, the OH density outside the tube increase slightly when the working gas is mixed with a small ratio of other gases (N$_2$, O$_2$ and air). Higher concentration of oxygen gas causes the dissociation of H$_2$O by O(1D) more vigorous, and it leads to a substantial increase in the OH density. Due to the effective diffusive gas mixing, the OH radical density increases only slightly outside the tube (Fig. 8).

Using helium as working gas with different purities (99, 99.99 and 99.9999%), the photoionisation of air species is found to play a crucial role in the transition from the localised discharge to streamer inside the tube. For the highest purity helium, the discharge plasma stopped inside the tube. The ionisation rate shown in Fig. 9 is concentrated in the head of plasma, however, the photoionisation shown in Fig. 9 is localised at the edge of the nozzle. This is because that the high concentration of air at the nozzle results in localised photoionisation, which cannot act as a

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**Fig. 10** He mole fraction of (a) 0.2 mm tube diameter and (b) 1 mm tube diameter. Plasma properties 84 ns after the start of pulse excitation of 0.2 mm tube diameter: (b) electron density; (c) ionisation rate. Plasma properties 103 ns after the start of pulse excitation of 1 mm tube diameter: (e) electron density; (f) ionisation rate. X-axis was the downstream direction of tube. The white dashed line in (d) suggests the position of the tube axis. +HV and small red block in (d) suggested the position of power electrode.

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**Fig. 11** Plasma properties

- a, d He$^*$ density
- b, e O$_2^*$ density
- c, f O density for 0.2 mm tube diameter and 1 mm diameter

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**Fig. 12** Pre-avalanche electron density at the same tube diameter

- a Ignition voltage and the pre-avalanche electron density for varying tube diameter. The pre-avalanche electron density inside the tube was obtained when the applied voltage was 2100 V for varying tube diameter. Two initial background electron densities 1 x 10$^{11}$ and 1 x 10$^{10}$/m$^3$ were used to analyse the effect of tube diameter on pre-avalanche electron density
- b Peak number density of He$, O_2^*$ and O at the position of 7 mm from the nozzle for varying tube diameter
source of pre-ionisation and provide enough electrons ahead of the streamer head. Therefore, the transition of discharge to streamer was not observed in this case and the plasma channel was truncated by the expanding sheath. When the purity of helium was decreased, the photoionisation extended to 1 mm ahead of streamer head, and the photoionisation rate was nearly two orders of magnitude higher than that with the highest purity. Therefore, the simulation results suggest that the photoionisation of air species acted as a source of pre-ionisation in front of the ionisation region.

3.2 Effect of tube diameter on an atmospheric-pressure micro-plasma jet

The 2D axisymmetric geometry is shown in Fig. 10. The computation range is 15 × 10 mm. The helium flow rate is 200 SCCM, and a pulsed direct current voltage with a rising phase of 50 ns is applied at a thin electrode embedded within the dielectric tube. The amplitude of voltage is from 2.6 to 4.3 kV when the range of tube inner diameter is from 0.1 to 1 mm. The vertical distance from power electrode to the tube inner surface is 1 mm while the horizontal distance between power electrode and nozzle of the tube is 2 mm. The electrostatic potential for the far-field boundaries of the ambient air region is grounded for all simulations presented (Fig. 10d).

The effect of tube diameter on sustaining voltage, electron density and the density of reactive species is investigated with the helium as the working gas. The numerical results proved that the ratio of the diameter of He–air mixing layer (helium concentration: 10%) and dielectric tube diameter is smaller as the wider tube diameter at the same gas flow rate. The peak value of electron density in the plasma bullet is higher as the tube diameter decreased. The density of active species also has a substantial increase when the tube diameter is narrow (Figs. 11a–f).

A systematic investigation of the connection between the ignition voltage and the tube diameter is presented, too. For the smaller dielectric tube, the drift movement of electrons to the inner wall leads to the depletion of electrons much more. More and more depletion of electrons then results in the decrease of the pre-avalanche electron density inside the tube. Therefore, to sustain the avalanche-to-streamer transition, the higher ignition voltage is necessary as the tube diameter decreased. In order to make the results more credible, we set different initial background electron density, and get the similar variation of the pre-avalanche electron density at the same tube diameter (Figs. 12a and b).

3.3 Simulations of plasma applications

3.3.1 Plasma treatment of bacteria biofilms: The schematic of geometry is shown in Fig. 13. The simulation geometry is 3 mm × 5 mm and the domain of bacteria biofilm is 0.1 mm × 0.4 mm are shown in Figs. 13b and c, respectively. The bacteria biofilms consist of several toe-shaped projections with cavities around them. The upper boundary is the metal electrode driven by

![Fig. 13](image_url) Schematic of geometry

- Schematic of the model geometry for DBD treatment of bacteria biofilm on the apple surface. Enlargement of the biofilm shows the structure of the bacteria biofilm on the cuticle and epidermic cells of the apple.
- Entire computation domain is 5 mm × 3 mm. Apple surface serves as floating electrode and the upper electrode is powered.
- Numbers 1–7 with different colours help readers to distinguish the fluxes to the surface of biofilm.

![Fig. 14](image_url) Reactive species densities inside the biofilm at 0.3 and 1 ns

- O density (1/m³)
- O³ density (1/m³)
- NO density (1/m³)
- H₂O₂ density (1/m³)

Maximum magnitude is indicated in each frame.

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pulsed DC voltage, the amplitude is $-25$ kV with 1 kHz pulse frequency and 1 ns pulse width. The thickness of dielectric barrier ($\varepsilon_r = 4$) below the metal electrode is 1 mm. The distance between power electrode and apple surface is 4 mm. The boundary at the bottom of the computational domain is grounded, so the air plasma is generated between the dielectric barrier and apple surface which serve as a floating electrode in experiment.

The simulation results suggest that the distribution of ROS and RNS is related to the structure of biofilm and the electrical parameters. The mean free path of charged species in $\mu$m scale determines the plasma can still penetrate into the cavity of the biofilm, although the non-uniform distribution of ROS and RNS caused by the structure of bacteria biofilm which is mushroom-shaped. In pulse off period, the diffusion results in the uniform distribution of ROS and RNS (Figs. 16a–f).

The treatment of three bacteria biofilms by two streamers is also simulated for there are a few bacteria biofilms on the fruits, and the air DBD always consists of a large number of streamers. The

![Fig. 15](image1)

**Fig. 15** Fluxes of $O$ (a), $O_{1d}$ (c), $O_3$ (e), $O_2^*$ (g), $H_2O_2$ (i) and NO (k) recorded at 0.3 and 1 ns for the negative streamer over the biofilm. Fluxes are recorded along the biofilm surfaces 1–7 and the corresponding colour as indicated in Fig. 13c. Time fluence of $O$ (b), $O_{1d}$ (d), $O_3$ (f), $O_2^*$ (h), $H_2O_2$ (j) and NO (l) for the streamer over the biofilm.

![Fig. 16](image2)

**Fig. 16** Density of $O$ (a), $O_{1d}$ (b), $O_3$ (c), $O_2^*$ (d), NO (e), $H_2O_2$ (f) near the biofilm at 1 ms. Diffusion make the distribution of these reactive species more uniform in the region close to the biofilm. Maximum magnitude is indicated in each frame.
results suggest that two streamers treat a larger area, and three biofilms are simultaneously treated besides of the reactive species of one streamer case are higher than those of two streamers case (Figs. 17a–e).

Fig. 17  Electron density (a), O density (b), NO density (c), ionisation rate (d), O3 density (e) and H2O2 density at 1.0 ns for the case of two negative streamers over three bacteria biofilms on the surface of apples. The middle biofilm has the closest distance between a low-arc apple surface and the dielectric barrier among the three biofilms. Maximum magnitude is indicated in each frame.

3.3.2 Plasma treatment of cells immersed in culture media: The schematic diagram is shown in Fig. 18. Generally, many dissolved reactive species [ROS (O2, H2O2, HO2, O3) and RNS (O2NO, NO, NO2, NO3, O2NOH)] are produced by the delivery of plasma reactive species into the culture media, and the transport of O2NO and O2 is studied. The production of ONO2aq is dominated by rapid reaction between H2O2aq and

Fig. 18  Conceptual diagram of the simulation system. First model described the transport of plasma reactive species into culture medium. The second model described the intracellular concentration and reactions of the plasma reactive species in target cells suspended in the stirred culture medium.

Fig. 19  Time and space evolution of dissolved O2NO and O2− concentration

a  1 min plasma treatment
b  2 min plasma treatment
c  3 min plasma treatment, in the 2 mm culture medium
d  Time evolution of dissolved O2− concentration during 5 min standing after 3 min plasma treatment
Plasma treatment increased the peak concentration of ONO$_{2}^{−}$aq. The diffusion increased the concentration of ONO$_{2}^{−}$aq in the deep region (Figs. 19a–c) during no plasma treatment. This study also found that the main source for O$_{2}^{−}$aq is the attachment reaction of electrons − dissolved O$_{2}$, and the transport of O$_{2}^{−}$aq is similar to ONO$_{2}^{−}$aq (Fig. 19d).

The pH of the liquid has significant effects on the concentration of O$_{2}^{−}$, HO$_{2}$, O$_{2}$NO$^{−}$ and O$_{2}$NOH in the culture media. When the solution pH was 4.5, the superoxides converted to hydroperoxyl radicals, and HO$_{2}$ is increased by the reaction between superoxide and hydroperoxyl radicals (O$_{2}^{−}$ + H$^{+}$ → HO$_{2}$). The influence of pH value of culture media on ONO$_{2}^{−}$ and ONO$_{2}^{−}$ concentration is shown in Fig. 20. With the pH value decreased from 7 to 4, the concentration of H$^{+}$ is increased rapidly, the ONO$_{2}^{−}$ concentration is decreased so quickly, and the ONO$_{2}^{−}$H production is dominated by the attachment reaction H$^{+}$ + ONO$_{2}^{−}$ → ONO$_{2}^{−}$H. The biological effect of peroxynitrite species is more significant with higher concentration of ONO$_{2}^{−}$H converted by ONO$_{2}^{−}$aq.

The effect of cellular plasma reactive species on the pH of the liquid is also sensitive. When the pH of culture media decreased to 4.5, the concentration of HO$_{2}$ in the cytosol increased (Figs. 21a and b). The fast increasing concentration of superoxide leads to attack cytosolic targets of captive cells [56], therefore, the bacteria are efficiently inactivated under acid condition [36]. The intracellular concentrations of ONO$_{2}^{−}$ and ONO$_{2}^{−}$H are also increased (Figs. 22a and b).

4 Conclusion

In this paper, the propagation mechanism of plasma and some plasma applications models are reviewed. First, the key equations related to plasma physics and chemistry are presented, including plasma governing equations, three-term exponential Helmholtz equations, plasma chemical reaction equations, and the Navier–Stokes, continuity and convection–diffusion equations. The computational model of the interaction between the plasma and cells immersed in the culture media is also introduced. Second, 2D simulation model finds that the sheath forms near the dielectric tube inner surface, which results in the plasma channel to shrink in the radial direction inside the dielectric tube. The photoionisation of air species plays a crucial role in the transition from the localised discharge to streamer. The Penning ionisation increases the electric conductivity of the plasma channel and facilitates the formation of ring-shaped plasma bullet. For the plasma jet in the open air, electron-impact dissociation of H$_{2}$O, electron neutralisation of H$_{2}$O$^{+}$, as well as dissociation of H$_{2}$O by O(1D)

Fig. 20 Stable concentration of O$_{2}$NO$^{−}$aq and O$_{2}$NOH$_{aq}$ (x > 1.5 mm) at eighth minute of 3 min/5 min in the culture media with different pH values

Fig. 21 Intracellular concentrations of O$_{2}$NO$^{−}$ and HO$_{2}$ in the cytosol

a O$_{2}$NO$^{−}$ concentration in the cytosol (r 0–10 μm) and in the film (r 10–20 μm) region. Legend 1 min/5 min means 5 min after 1 min plasma treatment. Legend 3 min/1 min means 3 min after 1 min plasma treatment

b HO$_{2}$ concentration in the cytosol (0–10 μm) and in the film (10–20 μm) region. The legend without mentioning pH means its pH = 7

Fig. 22 Intracellular concentrations of O$_{2}$NO and ONO$_{2}H$

a Effect of pH value of culture media on O$_{2}$NO$^{−}$ concentration (3 min/5 min) in the cytosol (r 0–10 μm) and in the film (r 10–20 μm) region

b Effect of pH value of culture media on ONO$_{2}H$ concentration (3 min/5 min) in the cytosol (r 0–10 μm) and in the film (r 10–20 μm) region
are found to be the main reactions to generate the OH species. For micro-plasma jet, the higher ignition voltage as the tube diameter decreases is attributed to the decreasing pre-avalanche electron density inside the tube. Finally, some latest simulation advances such as plasma treatment of bacteria biofilm, plasma treatment of cells immersed in liquid are presented.

5 References

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