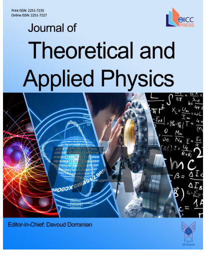

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Simulation of the impact of humidity on the species generated by a one-dimensional discharge of helium gas

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Original Research

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Abstract:
Dielectric barrier discharge (DBD) plasma has several applications in different fields. One of these fundamental applications is medical usages, where various methods are employed to improve the plasma treatment process. The combination of different gases is one of the important strategies to improve the performance of plasma in treatment. In this paper, the optimized plasma parameters for one-dimensional radiofrequency discharge produced at low pressures in a helium gas combination is studied. In this research, the optimal combination of H_2O and He is identified to attain the highest amount of reactive oxygen species (ROS). Considered mixture are 5, 10, 15 and 20 precent of H_2O for one dimensional helium gas discharge. The results show that the parameters of the output plasma are highly dependent on the composition of the input gases. It is found that the greatest concentrations of H^+, He^+, Hes (excited helium), and OH densities are observed when the H_2O percentage was at 10%. Moreover, the density distributions of various species and the temperatures of electrons are numerically calculated during the electrical discharge process. These findings provide useful knowledge on how to optimize plasma parameters for biomedical applications, which may lead to improved treatment results in several therapeutic areas.

Keywords: Cold plasma; DBD; Gas mixture; Plasma simulation; Plasma medicine

1. Introduction

The plasma medicine is a relatively recent issue which offers novel approaches to treat a wide range of illnesses. Due to the unusual quality and extended range of medical applications, including bio-sterilization [1], skin regeneration [2], wound healing [3], teeth whitening [4], blood coagulation, cancer cells treatment [5–7], and engineering of biomaterials and tissues [8], cold plasma discharge produced at atmospheric pressure has paid more attention in recent years. Generally, cells, tissues, and organs can all be treated employing the so-called cold atmospheric plasmas. The term "cold" describes a crucial characteristic of particular types of plasma, such as the fact that the temperature of the ions and other heavy species present in the plasma is much lower than that of the plasma's electrons [9, 10].

Dielectric barrier discharge (DBD) offers a higher intensity, more flexible, and regulated discharge in comparison to other plasma sources which is used by the majority of biomedical devices utilizing in the cold plasma discharge [11]. A lot of literatures has been focused on identifying and analyzing the plasma properties and active species during performing successful experimental research on atmospheric pressure plasma applications in surface treatment, engineering [12, 13], processing technology, and sterilizing [14, 15]. Additionally, different research groups have studied on simulating DBD plasmas at atmospheric pressure. For instance, Gadkari et al. [16] used a 2-D fluid model in COMSOL Multiphysics to simulate a co-axial DBD plasma reactor in pure helium. They examined how partial packing affected the helium dielectric barrier discharge's properties.

In addition, Pan et al. [17] numerically studied on different features of the atmospheric-pressure CF_4 plasma in a dielectric barrier discharge using the fluid model. They obtained the plasma parameters in steady state. Furthermore, Abidat et al. [18] investigated a one-dimensional model of atmospheric pressure helium gas dielectric barrier discharge with COMSOL Multiphysics simulation. They reported the effect of dielectric coefficient and distance between electrodes on Lissajour image. Golubovskii et al. [19] used the numerical methods to study the spatiotemporal properties of the homogeneous barrier discharge in helium. They utilized a one-dimensional fluid model to investigate how the main processes affect the discharge via rate constants. Moreover, they extracted the plasma parameters in steady state.
Enterally, combining gases is one of the key methods to enhance the plasma performance in medical applications [20–22]. This is because using different gas combinations can produce other active species that are beneficial for medical applications. Employing various types of gases with different compositions is one of the ever-toughest techniques in plasma modeling. Therefore, in this study, a simulation is performed for exploring of inlet gas combination portion in cold plasma. Different portions of H_2O are mixed with helium gas and plasma characteristics had been investigated. As a result, the best and effective combination of these two gases was determined for medical applications.

2. Theoretical equations

2.1 DBD simulations in sterilization process

In this section, the governing fluid dynamic equations in the DBD simulation for sterilization purpose are described. In order to account for the reactions of various species, and as well as the rate of production and losses at the electrode surfaces, surface chemistry was applied [23]. The pair of propulsion and propagation equations were solved to compute the electron density and electron mean energy. The electron continuity equation and the flow equation can be explained by Equations (1) and (2), respectively as below:

$$\frac{\partial n_e}{\partial t} + \nabla \cdot \mathbf{\Gamma}_e = R_e - (\mathbf{u}.\nabla) n_e \tag{1}$$

$$\mathbf{\Gamma}_e = -(\boldsymbol{\mu}_e \cdot \mathbf{E}) n_e - \mathbf{D}_e \cdot \nabla n_e \tag{2}$$

where, n_e represents the electron density, E is electric field, D_e indicates the electron diffusion coefficient, Γ_e is electron flux, u is average species fluid velocity, and R_e represent the rate of electron creation.
It should be mentioned that two terms construct the electron flux: i) one term is originated from the electric field, and ii) the other is created from the density gradient. Equation governing on the electron energy density can be calculated by:

$$\frac{\partial n_\varepsilon}{\partial t} + \nabla.\mathbf{\Gamma}_\varepsilon + \mathbf{E}.\mathbf{\Gamma}_\varepsilon = R_\varepsilon - (\mathbf{u}.\nabla) n_\varepsilon \tag{3}$$

$$\mathbf{\Gamma}_\varepsilon = -(\boldsymbol{\mu}_\varepsilon.\mathbf{E}) n_\varepsilon - \mathbf{D}_\varepsilon.\nabla n_\varepsilon \tag{4}$$

where the energy received by the electron from the electric field is denoted by the $\mathbf{E}.\mathbf{\Gamma}_\varepsilon$. In addition, the energy rate resulting from inelastic collisions can be expressed by the following equations:

$$R_\varepsilon = S_{en} + \frac{Q + Q_{gen}}{q} \tag{5}$$

$$D_\varepsilon = \mu_\varepsilon T D_e = \mu_e T_e \mu_\varepsilon = \frac{5}{3}\mu_e \tag{6}$$

In above equation, u_e is the energy mobility, and Q_{gen} is the thermal source, S_e is the power dissipation, Q is the primary source of heat, and q is the electron charge. It should be mentioned that due to knowing that the electron source could be determined from the Townsend coefficients must be used as:

$$R_e = \sum_{j=1}^{M} x_j a_j N_n |\Gamma_e| \tag{7}$$

where, M is the number of reactions, x_j is the molar percentage of the target species for the reaction j, a_j is the Townsend coefficient for the j process, and N_n is the total number of the neutral species considering that p is the number of non-elastic collisions of an electron.

$$R_\varepsilon = \sum_{j=1}^{p} x_j a_j N_n |\Gamma_e| \Delta\varepsilon_j \tag{8}$$

here, $\Delta\varepsilon_j$ represents the energy released through the j reaction. Generally, for each mass fraction in non-electron species, the following equations must be solved:

$$\rho \frac{\partial w_k}{\partial t} + \rho(\mathbf{u}.\nabla) w_k = \nabla.\mathbf{J}_k + R_k \tag{9}$$

where, w_k is the ion density and J_k is the ion energy flow. The Equation (10) can be utilized for determining the electrostatic field as follows:

$$\nabla(\varepsilon_0 \varepsilon_r \mathbf{E}) = \rho \tag{10}$$

where, ε_r is the relative dielectric constant and ε_0 is the permittivity of the vacuum. The following equations explains the boundary conditions for electron flux and energy flux as bellows:

$$-\hat{n}.\mathbf{\Gamma}_e = (\frac{1}{2}\upsilon_{eth} n_e) - \sum_p \gamma_p (\mathbf{\Gamma}_p.\hat{n}) \tag{11}$$

$$-\hat{n}.\mathbf{\Gamma}_\varepsilon = (\frac{5}{6}\upsilon_{eth} n_e) - \sum_p \varepsilon_p \gamma_p (\mathbf{\Gamma}_p.\hat{n}) \tag{12}$$

The electron is produced according to secondary emission, as shown by the second term on the right side of Eq. (11). Here, Γ_p is the secondary emission coefficient. It should be noted that the second term in Equation (12) is the secondary emission energy flux, where ε_p is the mean energy of the secondary electrons.
Ions and excited species are neutralized particles on the electrode surface through the surface reactions. The coefficient β_j which determines the probability of j species functioning is used to imitate the surface interactions on the electrodes. The conjugation equation for the discharge's ion species is expressed as follows:

$$\frac{\partial n_i}{\partial t} + \nabla.(n_i \mathbf{u}) = -\nabla.(\mu_i n_i q_i \nabla\varphi - D_i \nabla n_i) + S_i \tag{13}$$

where, φ and S_i show the electrostatic potential and the rate of variation of electron density, respectively.

2.2 Chemical model

This model incorporates the ground-state species of helium atoms (He), steam molecules (H_2O), OH molecules, and hydrogen atoms (H). Additionally, excited species above the ground state level have been taken into account. These include helium atoms (Hes), steam molecules (H_2Os) and OH molecules (OHs), respectively. Moreover, helium ions (He^+), H_2O ions (H_2O^+), hydrogen positive ions (H^+), OH positive ions (OH^+), Oxygen atom (O) and electrons (e) have been considered. These species are extremely important due to their presence and applications in He-H_2O plasma which has been discussed in experimental works [24–27]. The different species included in the plasma model are listed in Table 1. In total, 14 plasma species and 19 distinct processes are all involved in the He–H_2O mixture. In this model, Tables 2 and 3 provide an extensive overview of the possible reactions that were taken into consideration for helium and H_2O species.

Generally, surface interactions enable heavy species, such as positive ions, atoms, and metastable atoms to be transferred to the reactor wall. Since negative ions are unable to escape from the ambipolar field or reach to the reactor walls, no surface reactions are required. It is noticed that the only way to extract negative ions from the plasma is interaction with positive ions. This model includes the surface reactions listed in Table 4. Moreover, the decay coefficient rate for neutral particles to the wall surface is calculated using the sticking coefficients.

2.3 Boundary conditions and reactions

In this research, the initial magnitude of the electron density in the plasma is constant equal to 1×10^{13} m^{-3}, and the densities of neutral species of helium and water molecules are initially considered constant at fractional scales. In addition, the temperature of the gas is considered equal to the room temperature i.e., 300 K. The initial electric potential is in ground state condition i.e., as 0 V, and the initial mean electron energy throughout the whole domain is considered as 5 V. Moreover, the discharge is powered by a Sino negative voltage power source at 750 V.

Here, a dimensional axisymmetric model is utilized to describe the electrical and energetic characteristics of the RF discharge in the He/H_2O combination. The finite element method is employed for calculating different plasma parameters.

In this research, the simulation's material elements are defined in the air medium by considering the species' pertinent reactions. The Boltzmann's equations are solved and using the cross-sections from the LXCAT data source [28], rate coefficients were determined. The reaction rates are extracted from different references [24, 25] as mentioned in above tables. One of the electrodes is used in this insulation simulation, and the other one is the same of that.

Here, the electrodes are circular plates with a diameter of 0.1 m. All sections have a predetermined spacing of 2×10^{-4} m between the electrode and the insulation, which is the empty area where plasma production occurs. The computational environment for implementing one-dimensional reactor geometry and the related meshes is depicted in Figure 1. Here, the dielectric distance is 1×10^{-4} m. For the meshing of the discharge space, a mesh with a symmetrical distribution with element number of 200 and rate of 5 is used. Moreover, for the dielectric space, it is selected from the predefined (extremely fine) mesh with maximum element size of 3×10^{-6} m.

3. Results and discussion

As referred in different [29, 30], the electron density and its temperature have a significant impact on medical applications. Therefore, in this paper, the main purpose is finding the optimized plasma parameters for utilizing in biomedical surface sterilization. Here, the plasma is a radiofrequency discharge operating at 1 atm pressure. Furthermore, about 20% of the gas combination is made from H_2O. As a first step, the evolution of the electric potential as a function of distance is shown in Figure 2, in the various percentages of He + H_2O mixtures like: He + 20% H_2O, He + 15% H_2O, He + 10% H_2O, and He + 5% H_2O at 0.0015 s. As it is clearly seen in this figure, a concavity is observed in 0. Moreover, the highest magnitudes of electric field are related to He + 5% H_2O mixture.

In addition, spatial distribution of electron temperature is presented in Figure 3 as a function of different percentages of H_2O (5, 10, 15, and 20%). As it is obviously seen in Figure 3, the maximum magnitude of temperature is happened at sh = 0.1. Moreover, at lower positions, a minimum is observed especially at 0.05, but it will be vice versa at higher positions or near to electrode.

A comparison among various voltages and currents of different percentages of H_2O mixture in He gas is presented in Figure 4. As shown in this figure, an oscillational behavior is appeared for voltage variations. According to the Figure 4, the lowest electric current is obtained at the humidity level of 0.1, which can be caused by more electron collisions at this humidity level, where fewer electrons reach the opposite surface and less current is obtained. These collisions lead to the production of electrons and the density of more positive species. The accumulation of heavy species

Table 1. Various species incorporated in the plasma model simulation.

Neutral Species	Exited Species	Ions	Electrons
He	Hes	He^+	e^-
H_2O	H_2Os	H_2O^+	
OH	OHs	OH^+	
H		H^+	
O		O^+	

Table 2. List of the main reactions including elastic, excitation, and ionization phenomena [31].

Reaction	Formula	Type	$\Delta\varepsilon$ (eV)
1	e+He→e+He	Elastic	0.00
2	e+He→e+Hes	Excitation	19.80
3	e+He→2e+He^+	Ionization	24.60
4	e+H_2O→e+H_2O	Elastic	3.04×10^{-5}
5	e+H_2O→e+H_2Os	Excitation	4.59×10^{-1}
6	e+H_2O→2e+H_2O^+	Ionization	0.13

Table 3. List of some reactions with He specie by insertion of rate coefficient [31].

Reaction	Formula	$K^f f(m^3/s.mol)$
1	Hes+Hes→e+He+He^+	1.50×10^{-16}
2	Hes+2He→He_2s+He	2×10^{-46}
3	H_2O+He^+ →He+OH+H^+	2.04×10^{-16}
4	H_2O+He^+ →He+H+OH^+	2.86×10^{-16}
5	H_2O+He^+ →He+H_2O^+	6.05×10^{-17}
6	H_2O+Hes→O+2H+He	1.0×10^{-16}
7	O+Hes→He+O^++e	4.3×10^{-16}

Table 4. Surface reactions with insertion of the sticking coefficient [32].

Reaction	Formula	Sticking Coefficient
1	He^+ →He	1
2	Hes→He	1
3	H_2O^+ →H_2O	1
4	H_2Os→H_2O	1
5	OH^+ →OH	1
6	H^+ →H	1

causes the deviation of electrons and causes fewer electrons to reach the opposite plane.
Figure 5 represents the Root Mean Square (RMS) variations of the electron density for different mixtures of H_2O in He gas as a function of distance. A similar trend in magnitudes as seen in Figure 3 can be observed in Figure 5 too. On the other hand, the greatest values in the electron density occur for 0.1 of H_2O similar to the case of the maximum magnitudes of the electron temperature. In all mixtures, after $x = 5.0$E-5, a growth in electron densities is presented, while a downward trend is shown near to electrode surface. Moreover, a minimum in density is observed for whole H_2O mixture except to 0.1.
The RMS distribution of H^+ density for various H_2O mixtures in the He gas are shown in Figure 6 as a function of distance. As it is seen in this figure, the greatest magni-

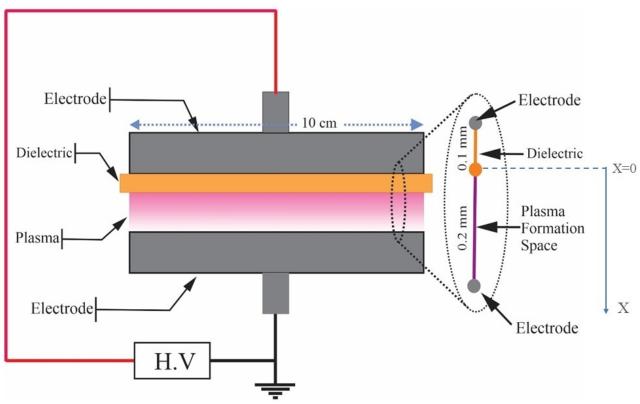

Figure 1. A schematic of the structure of the plasma by presenting the locations of the electrodes on the surface and inside of the behind dielectric.

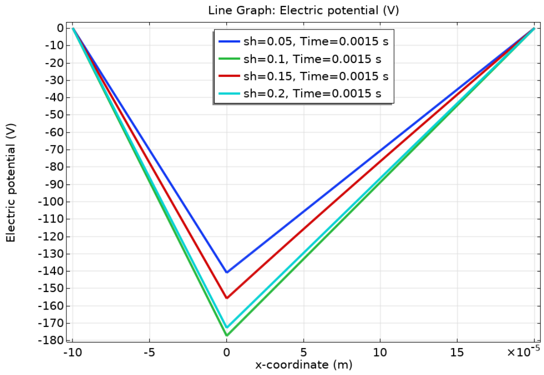

Figure 2. The evolution of the electric potential versus distance, for various percentages of H_2O mixtures of 0.05, 0.10, 0.15, and 0.20.

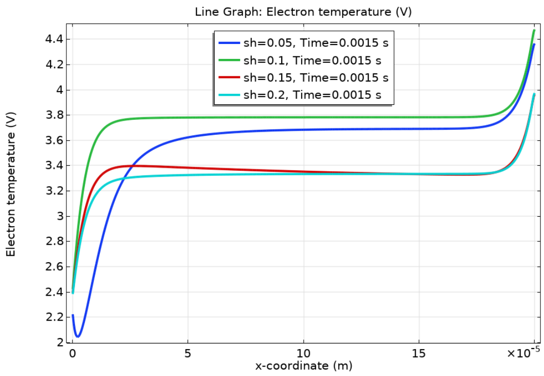

Figure 3. The spatial distribution of the electron temperature at 0.0015 s, for different percentages of H_2O at sh = 0.05, 0.1, 0.15, and 0.2.

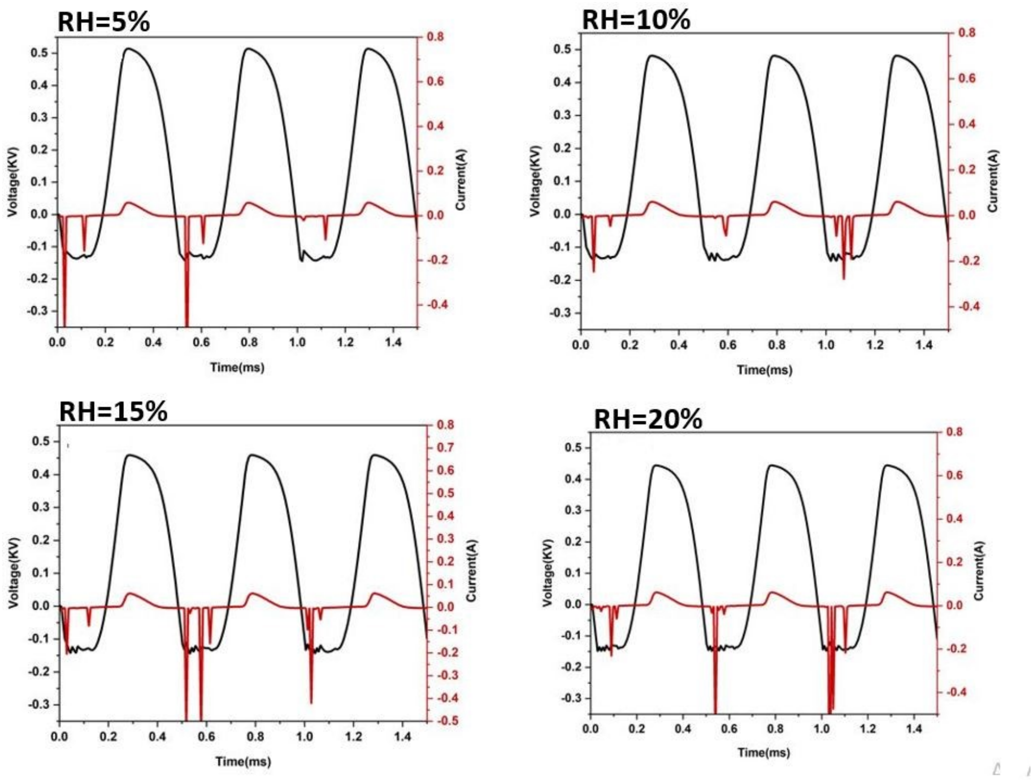

Figure 4. The evolution of voltage and current versus time, for different percentages of H_2O.

tude of H^+ density occurs at 0.1 H_2O and then, a sudden downward trend is observed in this concentration. Furthermore, for two mixtures of 0.1 and 0.15, a decrease in H^+ densities are observed in $x = 2E\text{-}4$, i.e. in the vicinity of the electrode surface. The numerical value obtained for the electron density for an atmospheric pressure plasma is in good agreement with previous simulation and experimental works [33, 34].

In Figure 7, the evolution of the H_2O^+ density is presented as a function of distance. As it is shown in this figure, the H_2O^+ density for 0.1 mixture has a downward trend. Figure 7 also shows a similar behavior in magnitude to that of Figure 6. As it is seen in this figure, the other percentages represent approximately constant magnitudes with respect to the distance from the electrodes.

The spatial evolution of He^+ density is illustrated in Figure 8 which represents the highest magnitude for 0.1 mixture. Furthermore, at 0.1 mixture, first an increasing trend is seen, but decreasing trend is observed at 2.0E-4. For two percentages of 5% and 20%, a concavity appears at an approximately distance of 1.0E-4 from the dielectric, while for the 15%, the minimum is occurred at 5.0E-5.

The density of excited helium is illustrated in Figure 9. Clearly, at 0.1 H_2O mixture, with highest magnitude show an upward trend between two electrodes. In addition, for all combinations, a drop is presented near the electrode surfaces at $x = 0$ and 2E-4 m. Moreover, at a distance of 0.75E-4 from the dielectric, a concavity is presented for two percentages of 5 and 20%.

OH radicals have been reported to enhance chemical processes [35] and can cause damage to the fatty acid side chains of lipids in different membranes, including the mitochondrial membranes of cells [36]. Figure 10 demonstrates the variations of OH density as function of x coordinate. In this figure for percentage of H_2O 0.1 mixed, a downward trend is represented, especially at distance of 2E-4 from the first dielectric. In the other mixtures, a constant density is observed, while at 2E-4 meters from the dielectric, all the graphs converged.

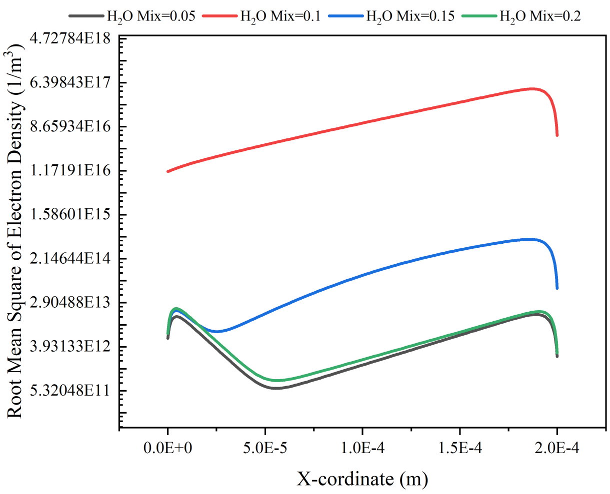

Figure 5. The spatial distribution of the electron density for different percentages of H_2O at sh = 0.05, 0.1, 0.15 and 0.2.

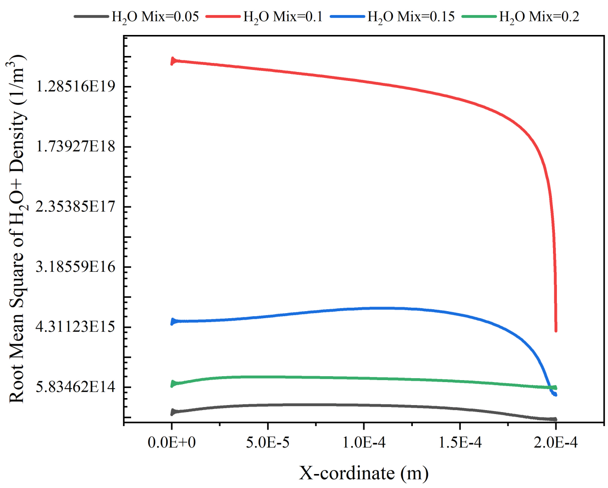

Figure 7. The regional distribution of the H_2O^+ density for different H_2O ratios for various mixtures of sh = 0.05, 0.1, 0.15, and 0.2.

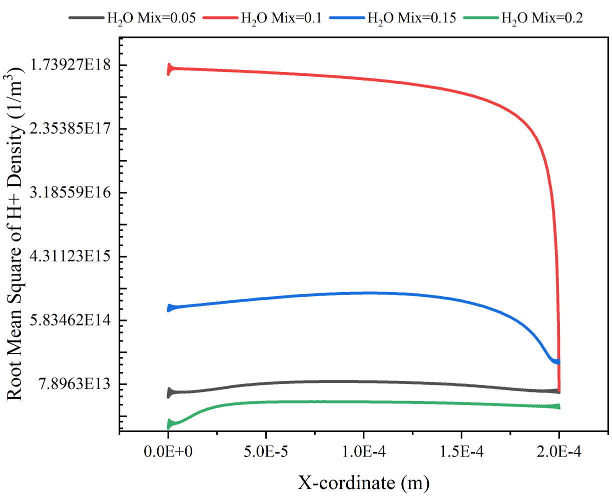

Figure 6. The spatial distribution of H^+ density for various H_2O percentages of sh = 0.05, 0.1, 0.15 and 0.2.

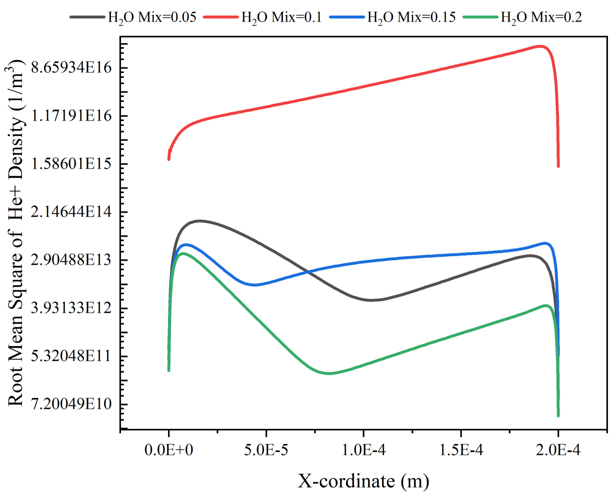

Figure 8. The evolution of He^+ density versus distance, for different percentages of 0.05, 0.10, 0.15, and 0.20.

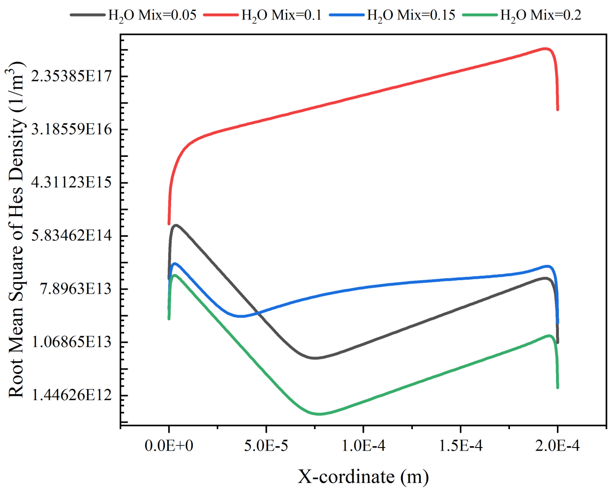

Figure 9. The spatial distribution of the exited helium density for different percentages of H_2O at sh = 0.05, 0.1, 0.15, and 0.2.

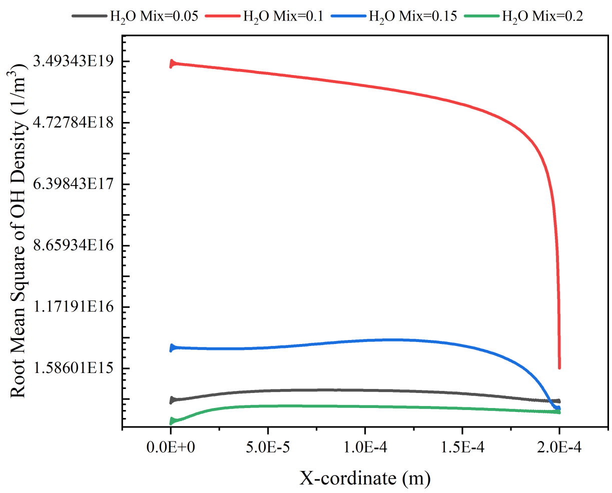

Figure 10. Variations of OH density as a function of distance, for different mixtures of H_2O with 0.05, 0.1, 0.15, and 0.2.

The existence of atomic O, an active species, in plasma-especially cold plasma-is crucial, particularly for applications in medicine. As shown in Figure 11. As the amount of moisture increases, the atomic number of oxygen increases, but this change is not very significant due to the lower contribution.
The amount obtained for the species was compared with previous works. Due to the fact that the plasmas utilized in the earlier studies were specifically tailored for various purposes, the parameters of the plasma, such as voltage, were set at greater levels, thus resulting in an increase in the discharge gap. However, the particle density is directly proportional to the atmospheric pressure plasma of helium-water gas [24, 34, 37].

4. Conclusion

In this paper, research was conducted on the optimized plasma parameters for the generation of radiofrequency discharge plasma under low pressures in a combination of

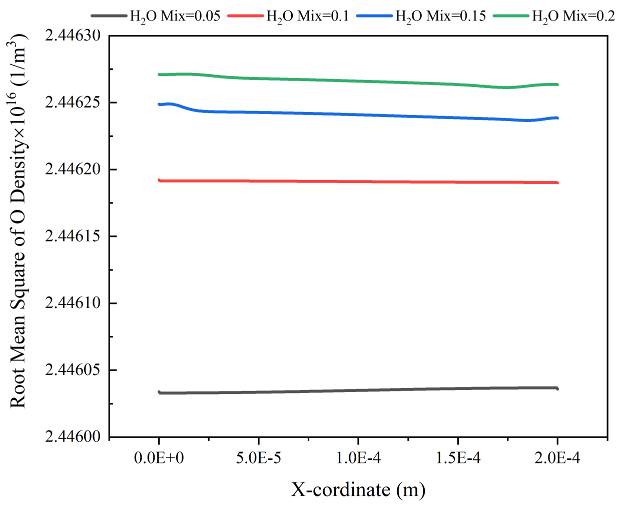

Figure 11. Variations of O density as a function of distance, for different mixtures of H_2O with 0.05, 0.1, 0.15, and 0.2.

helium. The main purpose was to investigate the potential of this plasma discharge in the field of biomedical surface sterilization. Through numerical calculations, the density distributions of various species and the temperatures of electrons during the electrical discharge process were determined. The results showed that the highest magnitudes of electric field were related to He + 5% H_2O mixture. Furthermore, it was shown that the maximum magnitude of electron temperature was happened at sh = 0.1 or percentage of He + 10% H_2O mixture. On the other hand, it was found that the highest magnitudes of voltage occurred at 0.05 mixture of H_2O. In addition, similar to the case of the maximum magnitudes of the electron temperature, it was seen that the greatest values in the electron density occurred for 10% percentage of H_2O. Moreover, it was found that the highest magnitudes of H^+, He^+, Hes (exited helium) and OH^+ densities occurred at 10% H2O percentage. Therefore, it was concluded that the best optimization plasma parameters may observe at He with 0.1% H_2O percentage.

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Authors Contributions
Ramin Mehrabifard conceived the presented idea. Zahra Soltani performed the simulation. Ramin Mehrabifard and Hamed Soltani completed the simulation. Zahra Soltani wrote the manuscript. Mohammad Mohsen Hatami, Fatemeh Rezaei and Ramin Mehrabifard edited the manuscript. Mohammad Mohsen Hatami and Fatemeh Rezaei supervised the findings of this work. All authors discussed the results and contributed to the final manuscript.

Availability of data and materials
Data presented in the manuscript are available via request.

Conflict of Interests
The author declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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