The efficacy of a gas shield in eliminating the effects of ambient conditions on the treatment of a liquid sample with a plasma jet

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Abstract: A shielding gas device can be employed to eliminate the effect of different ambient conditions, such as humidity, on the treatment of a liquid sample with a plasma jet. However, not as much attention has been given to its effectiveness in preventing the influence from the ambient atmosphere in different treatment setups. Here, we use a 2D-axisymmetric fluid flow model to computationally investigate the treatment of liquid in different well-sizes, for different atmospheric conditions.

Keywords: Non-thermal plasma, CFD modelling, gas shield, plasma-liquid interaction

1. Introduction

Atmospheric pressure plasma jets (APPJ) are a typical source of cold atmospheric plasma (CAP) used in plasma medicine research, where they are used for the treatment of cells or tissue, in both *in-vitro* and *in-vivo* experiments. An important example of such an APPJ is the kINPen®, a medically certified plasma jet that operates with argon as the feed gas and has been the subject of numerous studies [1]. Additionally, the use of APPJs is gaining attention in other fields, such as nitrogen fixation [2], or polymer treatment [3]. Often, like in the above-mentioned kINPen, a noble gas is operated as the feed gas. The desired, plasma-produced species, be it for e.g. cell treatment or for changing the surface properties of a polymer, are reactive oxygen and nitrogen species (RONS). These reactive species produced by a plasma jet can originate from two possible sources: either from admixtures or impurities in the feed gas, or from mixing of the effluent with the surrounding atmosphere. The latter makes the plasma-treatment with this setup very susceptible to the atmospheric conditions during the treatment, such as the relative humidity, which will determine how much water can diffuse into the active plasma zone, and will thus play a role in the RONS-cocktail produced and the reproducibility of the treatment. To prevent this, a shielding gas device can be employed [4]. Here a second, concentric gas flow surrounding the jet separates the effluent from the surrounding air. The composition of this shielding gas can be controlled, thus allowing control over the gasses in contact with the plasma jet effluent. Though research has been conducted regarding the effect of the shielding gas composition on the effluent chemistry and by extension on the treatment effect [5-8], less attention has been given to the effectiveness with which the gas shield prevents mixing of the APPJ effluent with the surrounding atmosphere, especially for different setups.

For this work, we adapted a computational 2D-axisymmetric model for the kINPen plasma jet above a liquid water surface [9], and expanded it to incorporate a shielding gas device. With this model, we investigated how varying atmospheric conditions, such as temperature and relative humidity, affect the conditions in the effluent and by extension the chemical treatment of the liquid substrate. This allows us to assess the effectiveness of the gas shield.

In addition, these simulations were performed for different treated substrates. Treatment of cells with an APPJ in plasma-medicine research is typically performed in well plates of varying sizes. The treatment of such a well, as opposed to a flat surface, inherently creates a backflow towards the jet outlet, of which the flow pattern logically depends on the well geometry. As such, this flow may in turn influence the gas shield flow pattern.

2. Computational details

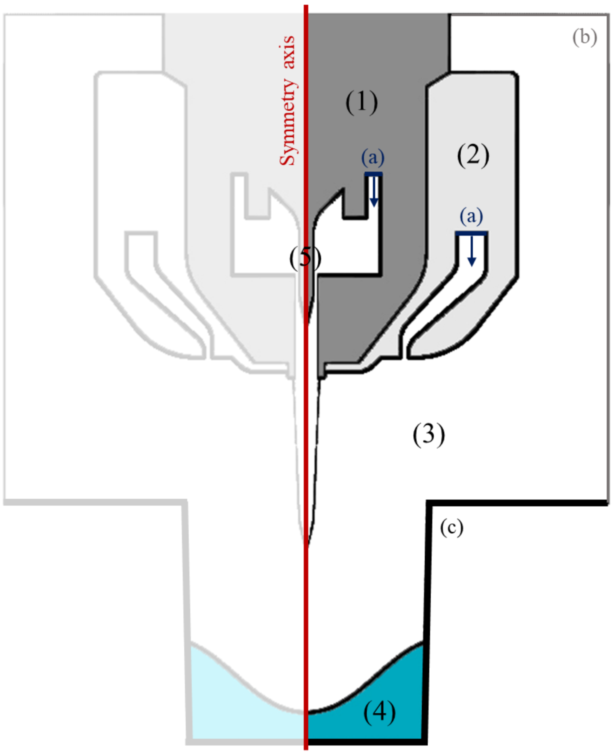

Fig. 1. General model geometry. The geometry components are (1) the plasma jet, (2) the shield gas device, (3) gas phase, (4) liquid phase and (5) the pin-electrode. Through boundary conditions the edges of the model are treated as (a) inlets, (b) open boundaries and (c) no-slip walls.

Figure 1 shows the model geometry of the plasma jet and shielding gas device, above a well from a 24-well plate. Treatment of wells from a 12-, 48- and 96-well plate was also investigated, all for a treatment distance of 2 cm, with 2 L/min of argon (containing ppm level impurities of O_2, N_2 and H_2O) flowing through the plasma jet, with and without a (4 L/min) gas shield. The shape of the liquid surface, induced by the gas flow impinging on it, was based on observations in our lab.

The model was built using the COMSOL Multiphysics software (version 6.0). The flow field in the system was calculated to a stationary state by solving the incompressible Reynolds-averaged Navier-Stokes (RANS) equations, employing the shear stress transport (SST) turbulence model in the gas phase. The liquid velocity field was solved as laminar flow. The calculated flow field was subsequently used in a time-dependent simulation for calculation of the conservation of energy (1) and the conservation of mass (2) in the system.

$$\rho C_p \frac{\partial T}{\partial t} + \nabla \cdot \vec{q} + \rho C_p \vec{u} \cdot \nabla T = Q \qquad (1)$$

$$\rho \frac{\partial \omega_i}{\partial t} + \nabla \cdot \vec{J_i} + \rho(\vec{u} \cdot \nabla)\omega_i = R_i \qquad (2)$$

Where ρ is the density, C_p the heat capacity at constant pressure, T the absolute temperature and ω_i the mass fraction of species i. $\vec{q}$ is the conductive heat flux, while $\vec{J_i}$ is the diffusive flux of species i, and $\vec{u}$ the velocity vector, implemented as the result of the stationary flow calculation described earlier. Q represents additional heat sources like, in this model, viscous dissipation and heat loss due to water evaporation. Finally, R_i represents the net production or consumption of chemical species (equal to zero in case no chemical reactions are included).

Heat transfer and chemical species transport were computed in a fully coupled manner, with species-specific properties, such as the diffusion constant and heat capacity, calculated within the model based on the mixture of species present at each time step. The model accounts for transport across the gas-liquid interface through Henry's law [10], and for water evaporation (including the resulting heat loss) through Antoine's law [11]. Turbulence in the flow field causes enhanced species transport, which is accounted for by implementing an additional diffusive term to the mass balance equation that depends on the turbulent viscosity. In this way, the mixing of the jet effluent with the surrounding atmosphere, as well as with the shielding gas, is simulated to investigate the conditions in the effluent that will in turn influence the chemistry and the liquid treatment. The plasma chemistry itself is not solved for in this 2D-axisymmentric model, but instead it is investigated in a quasi-1D model that utilizes the computational results from the 2D model. More details on this combined modelling approach can be found in [9].

The full model is applied for a range of ambient conditions including temperature (283 K – 303 K) and relative humidity (0% - 100%), as well as varying shielding gas compositions, to fully elucidate the effectiveness of the gas shield in eliminating environmental effects on the treatment.

3. Results and discussion

In literature, a gas shield has been reported a few times as a tool to experimentally mimic operation of the jet in ambient air, but with a controlled humidity [12, 13]. However, what these studies do not take into account is that the gas shield flow causes mixing with the plasma jet effluent which is significantly enhanced compared to mixing of the effluent with normal ambient air. Figure 2 shows the calculated molar concentration of nitrogen and oxygen gas in the centre of the jet effluent, as a function of distance from the pin electrode, both with and without gas shield. In both cases, the jet is surrounded by normal air (21% O_2, 79%N_2), coming from either the surrounding atmosphere or the shielding gas. Without a gas shield, as expected, the concentration of both ambient species rises gradually with distance from the pin electrode. In the presence of a shielding gas however, the concentrations rise much more rapidly, reaching values up to two orders of magnitude higher (cf. the logarithmic scale). This will significantly alter the RONS production in the afterglow.

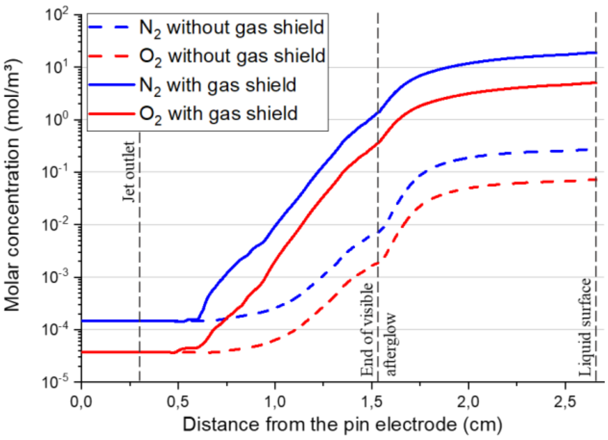

Fig. 2. Molar concentration of N_2 and O_2 on the symmetry axis, as a function of distance from the pin electrode, calculated for treatment of a 24-well both with and without gas shield with ambient conditions of 293 K and a relative humidity of 50%.

Regardless of the enhanced mixing, the gas shield is able to reduce variation in the jet effluent, e.g. water vapour concentration, that would be caused by different ambient conditions. However, the variations are not completely eliminated. In fact, we found that the main contributor to the remaining variation is the evaporated water from the treated well itself. Indeed, depending on the ambient temperature, more or less water will evaporate from the well during treatment.

Additionally, the substrate geometry determines the degree to which the gas shield can mitigate influence from the ambient. As shown in Figure 3(A), in case a 24-well is treated, the shielding gas envelopes the plasma jet effluent, reducing atmospheric influence up to an order of magnitude (as illustrated in Figure 3(C)). When a 48-well is treated however, the shielding gas hardly affects the jet effluent (see Figure 3(C)), and instead blows into the well next to that being treated as seen in Figure 3(B). This clearly indicates that in experimental research, the choice of the well size in which to treat the cells or the liquid can influence the treatment itself.

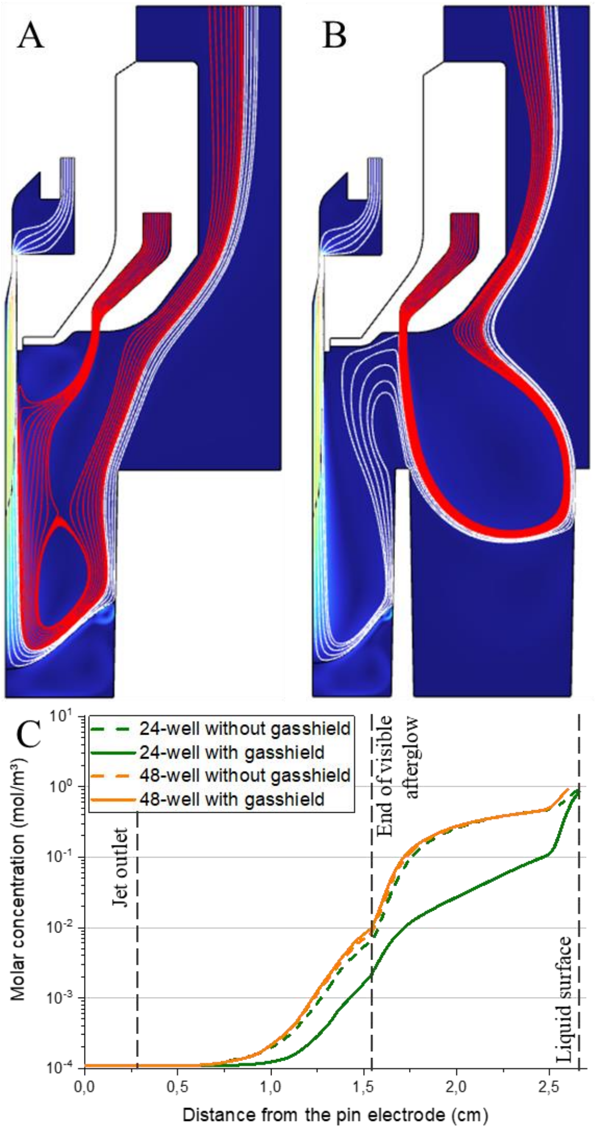

Fig. 3. A, B: streamlines of the flow field caused by the plasma jet (white) and shielding gas (red), with a 24-well plate (A) or a 48-well plate (B) as the treated substrate. In the latter case, the second well is also shown for clarity. C: molar concentration of H_2O on the symmetry axis, as a function of distance from the pin electrode, for treatment with and without gas shield, with ambient conditions of 293 K and a relative humidity of 50%.

Our computational results will be compared with experiments (results in progress), measuring the long-lived RONS concentrations in the liquid for various gas shield flow rates and compositions, and various well sizes, to validate our model predictions.

4. Conclusion

We used a 2D axisymmetric fluid flow model to investigate the influence of a shielding gas device on the treatment of a liquid in well plates of different sizes, with the kINPen® plasma jet, to elucidate its effectiveness in eliminating the influence from the ambient atmosphere. Our results indicate that even though the effects of the atmospheric conditions are reduced, they are not completely eliminated. The degree of shielding also depends on the geometry of the treated substrate. Our results also show that mixing of the jet effluent with the shielding gas is significantly higher than with the ambient when no gas shield is used. This must be taken into account in experiments, as it means that the gas shield cannot be used to mimic the surrounding atmosphere in a controlled way.

5. Acknowledgements

We acknowledge financial support from the Fund for Scientific Research (FWO) Flanders (Grant ID 1100421N, G033020N and 1SD6522N).

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