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A poly-diagnostic study of the shield gas-assisted atmospheric pressure plasma jet propagation upon a dielectric surface

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Abstract
The significance of shield gases on atmospheric pressure plasma jet (APPJ) propagation over a horizontal dielectric surface using various diagnostic methods has been investigated. The obtained results imply that adding nitrogen as a shield gas only has an impact on the N₂ reactive species generation, mainly close to the plasma effluent while argon shield gas can boost the optical emission intensity of all excited species, especially at distances further away from the plasma jet, which is in agreement with the observed length of the plasma propagation all over the surface. On the basis of the obtained results, the employment of a shield gas can improve the plasma jet efficiency to achieve the desired treatment effect on a flat surface.

KEYWORDS
atmospheric pressure plasma jets (APPJs), high-speed imaging, optical emission spectroscopies, plasma diagnostics, shield gases
1 | INTRODUCTION

The unique features of atmospheric pressure plasma jets (APPJs) have made them increasingly popular for plasma scientists over the past few decades. The enormous attention given to these devices is due to their intensified plasma chemistry as a result of the enhanced electron temperature while the plasma itself maintains a temperature close to room temperature. Furthermore, the long-term living species generated in the plasma jet and carried by the gas stream permit noncontact mode application. These unique features accompanied by having high flexibility, feasibility in the treatment of complex three-dimensional (3D) structures, and no requirement of expensive vacuum devices provoke a tremendous number of emerging novel APPJ applications and abundant investigations in different scientific domains such as, for example, biomedicine and material processing. Despite the ever-increasing utilization of APPJs due to their aforementioned excellent features, a higher performance of these plasma sources for practical purposes is still searched for. Many studies have been focused on the improvement of APPJ efficiency for specific applications. For instance, alteration of plasma processing parameters, such as treatment time, applied power, and gas flow rate, has been thoroughly investigated. Moreover, modifying the APPJ configuration, switching to a different discharge gas, changing the power source frequency, and controlling the ambient environment are some of the other examples of investigations performed to improve the effectiveness of APPJs. However, numerous challenges still exist to construct an APPJ source that can perform the required treatment with higher efficiency. In this study, the possibility of controlling the ambient species by introducing a shield gas into the APPJ effluent has been explored. Although this inexpensive method is applicable to all existing APPJ devices, only a few works have been committed to discovering the effects of a shield gas on the mechanisms responsible for plasma jet generation and propagation. In this context, it should be mentioned that manipulating the ambient environment plays a crucial role in improving the plasma reaction chemistry and avoiding unwanted species. This is of particular interest in different scientific fields such as the biomedical field as living cells are known to be more prone to respond to ambient impurities. As another example, during plasma jet treatment of miscellaneous objects, the effectively treated area can be simply tuned in the desired size and specific species can be incorporated, by a correct choice of the ambient conditions. Hence, for the sake of the previously mentioned purposes, an appropriate selection of a shield gas directed to the plasma effluent can be a cost-effective alternative to plasmas generated in closed low-pressure chambers. Aside from creating a controlled environment without the presence of unwanted species, a simplified approach for adding a shield gas has been introduced in this study in a way that the direction of the shield gas introduction can be controlled as desired. As such, the intersection of shield gases with the plasma gas stream can enhance point treatment even at remote distances allowing to treat a well-defined area by the plasma.

In an attempt to unravel the processes occurring during plasma formation and the plasma effluent development on a surface, diagnostic methods have been considered as a convenient tool. However, an in-depth diagnostic investigation necessitates the assessment of both the empirical view and simulation outcomes. Allegedly, gas flow dynamic simulations in combination with experimental observations facilitate drawing a vivid conclusion on the plasma propagation pattern. Previous studies were successful in the prediction of the plasma behavior and the surface treatment pattern, making use of this technique. Consequently, pursuing this approach provides comprehensive insights on the mixing of gases from different origin and their propagation all over a surface, which in turn reveal the processes responsible for plasma formation and extension over a surface. Accordingly, in this study, computational fluid dynamic (CFD) simulations of gas flows making use of Comsol MultiPhysics 5.3 were implemented to gather information on the velocity field and mass fraction of gas on a surface which enables us to reveal how the additional shield gases behave in contact with the discharge gas and a surface. As the plasma is yet another kind of fluid, a working APPJ can be seen as a fluids mixing process of plasma and ambient air. These fluids’ dynamics play a critical role in determining the plasma characteristics and define the plasma plume structure that reaches a sample surface. Schlieren imaging is an experimental technique that enables visualization of the changes in the refractive index caused by the presence of fluids with different densities for instance and assists with the interpretation of fluid-dynamic phenomena. Therefore, in this study, a fluid dynamic characterization utilizing Schlieren imaging of the plasma jet’s effluent region is also conducted. The combination of the qualitative flow depiction obtained from Schlieren imaging and the CFD simulation results can provide comprehensive insights on plasma propagation and its mixing with ambient air.

In the step after envisaging the gas flow behavior using simulations and imaging, the plasma jet properties should be identified using experimental diagnostic methods. Although a plasma jet driven by short pulses
(a few ns to a few µs) or with low excitation frequency (from a few Hz to a few kHz) at higher voltages is not a continuous discharge but rather consists of discrete structures which are named “plasma bullets.” Diagnostic techniques (such as optical emission spectroscopy [OES]) generally provide an integrated picture of the discharge due to the unmatching temporal resolution. However, it is critical to apprehend how each plasma bullet propagates in space and how it impacts the sample during its plasma exposure. These two combined observations yield a comprehensive overview of the plasma evolution in a specific area. Therefore, in this study, the plasma jet has been analyzed using OES as well as using a fast intensified charge-coupled device (ICCD) camera.

The current investigation intends to demonstrate a thorough perspective of the shield gas importance on APPJ characteristics based on the obtained findings by using multiple combinations of plasma processing parameters. CFD simulations, Schlieren imaging, OES results, and ICCD images all provide complementary constructive details on plasma jet spreading on a flat surface. The obtained results can open up a new level in the application effectiveness of an APPJ by exclusively modifying its ambient environment.

2 | EXPERIMENTAL SECTION

2.1 | Plasma jet set-up

In this study, a modified homemade APPJ, previously described and applied for the treatment of polymer surfaces was used. To introduce a stream of shield gas into the plasma effluent, small adaptations to the plasma jet were done, which will be described hereafter. First, three holes have been drilled in a polyvinyl chloride (PVC) disk (5 mm thickness and 60 mm diameter) at a 45° angle towards the plasma effluent and at a 10 mm radial distance from the capillary center in such a way that every single hole is equidistant from the two other holes. In the next step, three tubes of polyether polyurethane (inner diameter = 2.5 mm, outer diameter = 4 mm) were assembled into the holes and cut at the plasma jet side so that the tubes exactly end at the bottom side of the PVC disk. A single mass flow controller was used to deliver a specific shield gas into the plasma jet surrounding air. A schematic representation of the plasma set-up, together with a photograph of the working plasma jet, is presented in Figure 1. As the built plasma set-up is axially symmetric, a symmetry axis was chosen in this study to consider all possible conditions occurring due to the specific geometry. This symmetry axis covers two critical points which are interesting to examine: the first one is the point precisely underneath the plasma jet effluent, and the second one is the point between two tubes of shield gas (see Figure 1a). By doing so, the influence of the specific configuration of the designed set-up can be evaluated.

For igniting plasma, a 60-kHz high-voltage AC power supply with a fixed peak-to-peak voltage amplitude of 8 kV was used. A fixed flow rate of 1.0 standard liter per minute (slm) of argon (Air Liquide, Alphagaz 1) through the quartz capillary acting as discharge gas was used during all experiments. For the design of the experiment, different plasma operational parameters were selected to obtain a thorough insight into the relevance of the shield gas on the characteristics of the plasma afterglow. Nitrogen (Air Liquide, Alphagaz 1) and argon were chosen as shield gases in this study with overall flow rates of 1.5, 3.0, and 4.5 slm. This total flow rate was equally divided over the three channels of shield gas and thus resulted in 0.5, 1.0, and 1.5 slm flow rates into each tube, respectively. It is also worth mentioning that in this study the overall shield gas flow rate was used for identification and that the 0 slm shield gas flow rate was assigned to the experimental condition of no shield gas. Finally, for analyzing the resultant plasma with and without the shield gas, a PVC table with a square opening of 10 × 10 cm² in the middle to fit a quartz glass with 1 mm thickness was constructed (see Figure 1a). The distance between the edge of the capillary and this table was varied between three values: 5, 10, and 13 mm.

2.2 | Electrical characterization of the discharge

Measurements of the applied voltage and the discharge current were conducted to characterize the electrical behavior of the plasma. The voltage was measured with a high voltage probe (Tektronix P6015A) connected to the powered electrode, whereas a current transformer (Pearson Current Monitor Model 2877) was used to determine the current. The voltage–current waveforms were monitored with a digital oscilloscope (Picoscope 3204). On the basis of the obtained $I$–$V$ profiles, the average power ($W$) of the discharge was calculated according to the following equation (where $T$ is a single period of the discharge):

$$W = \frac{1}{T} \int_{t}^{t+T} I(t)V(t)dt. \quad (1)$$

Results are presented as an overlay of graphs obtained for argon and nitrogen shield gases with different gas flow rates and distances between the
capillary end and substrate to enhance data interpretation. All measurements were performed three times and the average power value is reported with a maximum ±0.2 W uncertainty.

### 2.3 CFD simulations

CFD modeling using Comsol MultiPhysics 5.3 was applied to gather information on the impact of the applied shield gas on the dynamics of the plasma jet main gas stream.[34] On the basis of the design of the experimental plasma set-up and the interaction of gases with the substrate (the quartz window), simulations were carried out under the assumption of turbulent flow.[16] Transport of concentrated species was used to gain information on the distribution of the mass fraction of argon atoms. Given the fact that there is no axial symmetry in the plasma set-up, a 3D geometry was applied for these simulations. Figure 2 represents the

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**FIGURE 1** (a) Schematic representation of the experimental set-up and (b) photograph of the working plasma jet with 3.0 slm argon shield gas and a 5 mm gap size.
constructed geometry of the plasma set-up in 3D view as well as in the YZ-plane view. Simulations were carried out in a cylindrical surrounding region with a 200 mm radius and a 50 mm height, which is assumed to be filled with air. While the main gas channel in the middle of the constructed geometry with a 0.65 mm radius played the role of the inlet for the discharge gas, three other tubes with 1.25 mm radius each were selected as shield gas inlets. The central channel corresponding to the discharge gas was fed by a fixed 1 slm argon flow while varying flow rates of nitrogen and argon shield gases were introduced in the other three inlets. The radius of the additional plate was 30 mm, which is identical to the plate used in the experimental set-up. Distances and gas flow rates were chosen as variables for computation of the mass fraction and the velocity field in the observation region. In these simulations, the incompressible flow was considered. The Navier-Stokes equation and the continuity equation were solved with a normal physics-controlled mesh in the volume of interest:

\[
\rho (u \times \nabla) u = -p I + (\mu + \mu_t (\nabla u + (\nabla u)^T)) + F, \tag{2}
\]

\[
\rho \nabla \times (u) = 0, \tag{3}
\]

where \(\rho\) is the density (kg/m\(^3\)), \(u\) is the velocity of a gas (m/s), \(I\) is the identity matrix, \(p\) is the gas pressure, \(\mu\) is the dynamic viscosity (Pa s), \(\mu_t\) is the turbulent dynamic viscosity, \(F\) is the volume force (N/m\(^3\)), and \(\nabla\) is the differential operator.

Transport of concentrated species was also introduced as follows:

\[
\nabla \times j_i + \rho (u \times \nabla) \omega_i = R_i, \tag{4}
\]

\[
N_i = j_i + \rho u \omega_i, \tag{5}
\]

where \(j_i\) is a relative mass flux component and \(\omega_i\) is a mass fraction.

### 2.4 Plasma flow visualization

A Schlieren imaging method was used in this study as it helps to visualize the difference in the fluid density through the refractive index changes in the medium. To observe the patterns of perturbation caused by the plasma flow and the shield gases, typical Z-type Schlieren imaging was employed in this study. Figure 3 demonstrates an example of this particular configuration which requires two concave mirrors, a light source, a sharp blade, and a camera.\(^{[35-37]}\) In this study, two parabolic mirrors (Edmund Optics) with f/10 aperture, 20.32 cm diameter, and 203.2 cm focal length were oriented at a 6° angle with respect to the horizontal axis and placed...
310 cm apart. As a point light source, a warm white (3000 K) light-emitting diode working in continuous mode was used. During the experiments, the plasma set-up was placed between the two mirrors in the parallel light beam, as shown in Figure 3. The camera (Nikon D3200) was focused on the plasma effluent, and photos were captured utilizing DigiCamControl software. A knife was mounted close to the camera lens to block the refocused light. Schlieren images were obtained for different experimental conditions to evaluate the impact of capillary-sample distance, shield gas flow rate, and the shield gas type on the fluid density in the volume between the extra PVC plate placed at the capillary edge and the quartz substrate.

It should be mentioned that during the Schlieren acquisition, each photograph for the same experimental condition is taken at different time steps which together with turbulence at a distance lead to small variations between each image. Thus, in this study, only one selected frame is reported for each experimental condition, which does, however, represent the image group’s general behavior.

2.5 | OES

OES was performed using an Ocean Optics S2000 spectrometer to collect information on the spatial distribution of the reactive species present in the plasma effluent and how this distribution was influenced by different experimental parameters such as shield gas flow rate, gap size, and shield gas type. An optical fiber was positioned perpendicularly and adjacent to the quartz plate placed underneath the plasma set-up and was moved along the symmetry axis, as defined in Figure 1a. Before starting the actual measurements, the complete optical system was calibrated using an Oriel model 63355 tungsten-D2 lamp. In the next step, OES spectra were recorded in the wavelength range 200–900 nm with a low resolution of 0.4 nm along the symmetry axis, and a 20-ms integration time was applied to obtain an appropriate signal/noise ratio for each studied OES line and band. The intensities of the acquired OES spectra are presented in arbitrary units.

2.6 | Light emission imaging

The propagation behavior of the plasma plume on the quartz window was visualized using two ICCD camera imaging techniques. In the first series of measurements, the dispersed emission pattern images of the argon plasma plume under different experimental conditions were obtained using a Hamamatsu ICCD camera (C8484). The exposure time was fixed to 500 ns for all imaging measurements, and the gain was kept constant at 50. These fixed settings allowed an objective comparison between the photographs obtained for all experimental conditions. The camera was placed underneath the experimental plasma set-up adjacent and perpendicularly to the quartz window to acquire sharp images of plasma propagation on the surface (see Figure 1a, ICCD camera 1). In the second series of measurements, the camera (4Picos; Stanford Computer Optics) direction was perpendicular to the jet effluent axis, as demonstrated in Figure 1a with ICCD camera 2. To observe the discharge development, a sequence of images was acquired using a shutter time of 100 ns (intensifier operates with maximum gain potential), while varying the trigger delay of the camera in steps of 100 ns. Each image demonstrates an accumulation of discharge events and, therefore, the sequence shows the average development of the plasma jet. The camera had an internal integration time of 16.7 ms, allowing it to accumulate all exposures up to 127 trigger events within this period before reading out the CCD sensor. The external trigger input was connected to
the trigger output of an oscilloscope, which displayed the voltage and current signals of the applied potential. The images obtained demonstrated the average intensity of 50 readouts, hence up to 6350 discharge events. For both set-ups no filter was used during imaging of the plasma propagation on the quartz surface; thus, the full ICCD spectral range of 290–900 nm was recorded.

3 | RESULTS AND DISCUSSION

The innovative APPJ design with the introduction of a shield gas as well as a partly trapped volume between the device and the substrate results in a plasma with unique features and these features will be thoroughly investigated in this section using a well-considered set of diagnostic techniques. More specifically, the impact of the shield gas on the plasma jet afterglow will be investigated by taking a closer look at variations in the discharge power, the intensity of reactive species, and the gas flow dynamics by means of CFD simulations and Schlieren imaging. Finally, the impact of the applied shield gas on the propagation of the plasma effluent on a flat surface will also be examined utilizing ICCD imaging.

3.1 | I–V characteristics

Using I–V waveforms, the electrical behavior of the discharge was studied. Figure 4 demonstrates an example of these waveforms with distinct periodic dynamics evident for the experimental set-up without any additional shield gas. It should be mentioned that no significant changes in the voltage–current waveforms were observed for all examined experimental conditions. The applied voltage has a sinusoidal shape while the discharge current reveals a displacement current combined with peaks in each half voltage cycle. These peaks are assigned to the plasma bullets, where the highest positive and negative charges are present. On the basis of the figure, in the positive half-cycle, the current peak initiates at a higher discharge voltage suggesting that the energy transferred to the plasma bullets in this half-cycle is higher. Therefore, the bullet propagation on the target occurs in the positive half-cycle. The average power (W) is determined based on the voltage–current waveforms using Equation (1) and the dependency of the calculated power values on the experimental conditions under study is shown in Figure 5. As shown in Figure 5, the obtained power values stay below 4.85 W for all experimental conditions with some changes in power values due to variations in the ambient conditions.

The first apparent result which can be retrieved from these curves is the influence of the shield gas type on the discharge power. When the same gas as the discharge gas (argon) is used as a shield, the discharge power is strongly enhanced as the combination of these gases helps the plasma to propagate over larger distances and to occupy a more significant volume between the additional PVC plate and the flat quartz surface. In this case, the complete plasma set-up resembles a plasma jet operating at a higher discharge gas flow rate which is known to result in a higher power when argon is used as the discharge gas. In contrast, the use of nitrogen as shield gas only has a negligible impact on the discharge power: it only slightly decreases the propagation of plasma by preventing the further development of the plasma.
plasma effluent on the surface due to mixing with nitrogen. This phenomenon will be later explained using OES measurements detecting different amounts of excited species for each experimental condition and will be additionally demonstrated by ICCD imaging while examining the plasma propagation on a dielectric surface with both shield gases under study.

Furthermore, the gap size (capillary–table distance) is another parameter causing variations in discharge power. As can be noticed in Figure 5, regardless of the shield gas type and its flow rate, an increase in gap size decreases the discharge power. Indeed, the proximity of the table prevents the dissipation of power in the volume between the PVC plate and the table, which consequently increases the discharge power at smaller distances. This increasing discharge power at smaller distances is very pronounced when using argon as shield gas at high flow rates. Indeed, the highest power value (4.85 W) is obtained when using a 4.5 slm argon shield gas flow rate and a 5 mm capillary–table distance. Another possible explanation for the increasing discharge power when reducing the gap size and adding argon as shield gas is the capacitance effect occurring in the volume between the additional PVC plate and the quartz window. The PVC disk, which in this geometry can be considered as a continuation of the capillary, is placed parallel to the table beneath while the plasma effluent propagates in the trapped volume between them. The capacitance of this particular configuration induces variation in the impedance of the complete system and subsequently increases the discharge power at smaller distances. This increasing capacitance effect occurs in a small gap results in the enhancement of the system capacitance, which directly increases the power.

### 3.2 CFD simulations

A fundamental understanding of how the different gases interact with each other and the surface can be obtained by investigating the propagation patterns of plasma on the surface with shield gas introduction. In our previous work,[16] the substantial impact of using an additional PVC plate on the velocity field and the argon mass fraction of a similar APPJ set-up has been investigated in detail. It has been shown that the extra plate constrains the gas in a closed volume and thus has a significant impact on the distribution of gases near the substrate. Accordingly, in this study, 3D numerical simulations were carried out by including three additional gas inlets for shield gas insertion to examine the plasma evolution on the surface and the volume above it. In the first step of the simulation, the distribution of gas velocities and their impingement on the surface were studied. It should be mentioned in this context that the velocity magnitude for both argon and nitrogen shield gas is following a similar distribution; thus, the essential features of velocity will only be illustrated using argon shield gas as an example. Figure 6 indicates the velocity field of argon for different capillary–table distances in the YZ-plane obtained for fixed main and shield gas flow rates of 1.0 and 4.5 slm, respectively. It is noteworthy to mention that the diameter of the main gas capillary and the shield gas tubes differ from each other (as previously mentioned) resulting in different gas speeds. The first apparent conclusion which can be drawn from Figure 6 is that the shield gas does not deviate from the main discharge gas stream to any particular direction. The second noticeable observation is that the magnitude of the shield gas velocity when it reaches the surface varies when using different capillary–table distances: the shield gas velocity at the surface is the highest in the case of a 5 mm gap size whereas for the 10 mm gap size (see Figure 6b) the impact of the shield gas on the surface is only moderate. In addition, for the 13 mm gap size, the shield gas velocity in the vicinity of the substrate is even lower. It is also worth mentioning that for the smallest distance between the capillary and the table, the argon shield gas streams from the three different tubes mix between themselves as well. The latter causes an extensive blend of the streams and the gases trapped in the small volume between the PVC plate and the horizontal quartz surface. The particularity of the experimental set-up geometry, which includes the position and inclination of the shield gas tubes, makes the 13 mm gap size distinct, as in this particular case, the shield gas simultaneously encounters the main discharge gas stream and the surface.

Another equally important parameter that influences the velocity field is the shield gas flow rate. Hence, such simulations were also performed, and the velocity distributions for a fixed 5 mm gap size and different shield gas flow rates are presented in Figure 7. When comparing Figure 7a with the others, it is evident that in the absence of a shield gas, a uniform distribution in velocity magnitude close to the surface emerges. However, adding a shield gas induces an uneven distribution in the velocity field and the highest disturbance occurs when a 4.5 slm shield gas flow rate is introduced. The lowest shield gas flow rate of 1.5 slm results in a stream that only has a negligible velocity when reaching the surface (see Figure 7b) whereas increasing the amount of injected shield gas produces a stream that actually reaches the surface (see Figure 7c,d).

The gap size between the additional PVC plate and the table is considered a critical working parameter that regulates the confined volume. Henceforth, the mass fraction of argon has also been examined for different distances between the end of the plasma jet capillary and
the table using a constant argon shield gas flow rate of 4.5 slm, and the outcomes are presented in Figures 8 and S1 in the YZ- and XY-plane, respectively. A closer look at the argon mass fraction distribution for the 5 mm gap size (Figure 8a) reveals the aggregation of argon atoms in a significant volume by the merging of the shield gas with the main argon discharge gas. These two gas sources provide a sufficient amount of gas to fill the small volume under the additional plate at high concentration, and consequently, the small volume under the plate can be treated as a supportive channel for further propagation of the plasma. In other words, the plasma jet will more
likely propagate in the direction where a higher concentration of the same gas is present. Figure 1b clearly visualizes the continuation of plasma propagation towards the shield gas direction. Thus, it is expected that in this study, under certain experimental conditions (small gap size and high flow rate of argon shield gas) the plasma will occupy a significant volume between the additional plate and the table. However, there is a limitation in the radial propagation of argon in high concentration, or plasma, due to the mixing with ambient air (see Figure S1a). By combining the simulation results shown in Figures 8a and S1a) it can be concluded that argon gas is present in high concentration in a small cylindrical volume under the additional plate, but its mass fraction is rapidly declining after the borders of the imaginary cylinder below the additional plate. At the same time, three argon-rich “tails” just outside the argon-rich cylindrical volume can be clearly observed in Figure S1a due to the continuation of the argon stream propagation in the direction of the shield gas injection. It is worth mentioning that if the whole volume would be completely filled with pure argon gas, the plasma would only slightly propagate more. Although in this case there is no interaction with air, the plasma energy only slightly survives longer. As the plasma species lifetime is connected to the source input energy, the propagation length will still be limited.

Simulation results for a gap size of 10 mm reveal that the main and shield gases are confluent as well although the mass fraction in their joining point is lower in comparison to a shorter distance between the additional plate and the table. The relatively smaller mass fraction of argon observed in the center of Figures 8b and S1b compared to the 5 mm gap size implies that increasing the capillary end-table distance reduces the concentration of argon thus weakening the supportive channel creation. However, in this case, the central volume still contains more than 75% of argon, which can still positively affect the resultant plasma propagation. Another notable difference between the results obtained for the 5 and 10 mm gap sizes is the distribution of argon on the table surface (see Figures S1a,b). However, for a shorter distance, a large area (almost circular) is covered with argon in high concentration, at medium distance, the shape resembles a concave triangle where argon has a higher mass fraction. In the latter case, three argon-rich “tails” are also formed by the continuous spreading of argon shield gas along the surface in the direction it is outflowing the volume between the additional plate and the table. Finally, totally different argon mass fraction patterns can be observed in Figures 8c and S1c) for the largest gap size. As mentioned before, the 13 mm gap size matches the distance where the gases encounter each other and the surface at the central point on the surface. Thus, considering the mixing between two argon streams and the substantial distance to the table surface, high dispersion of argon all over the volume close to the surface is detected. Notwithstanding the heterogeneity in
argon distribution in the volume between the plate and the table (Figures 8c and S1c) illustrates an excellent uniformity in spreading across the table surface with a set of argon-rich “tails” that are formed in the same manner as for the 10 mm gap size.

Another crucial parameter that has been evaluated in this study is the flow rate of the shield gas and the argon mass fraction simulation results obtained for a constant gap size of 5 mm are depicted in Figures 9 and S2. The top left corner of these figures shows the mass fraction of argon in the experimental set-up without the use of a shield gas flow. In this case, the only source of argon is through the plasma jet capillary, and the introduced gas fills the volume between the plate and the table almost uniformly. The backflow of argon gas inside the shield gas tubes can explain a small asymmetry in the argon distribution. Adding 1.5 slm argon shield gas to the system induces significant changes in the argon mass fraction distribution, which is shown in Figures 9b and S2b. As can be seen in these figures, the shield gas is not well incorporated into the main gas stream exiting the plasma jet capillary due to the difference in velocities, which was already discussed above. However, the shield gas does contribute to the pattern of the argon mass fraction distribution in the gap volume and on the table surface as a higher concentration of argon is mainly located close to the horizontal surface and argon spreads on the table in three directions corresponding to the shield gas injection. On the contrary, as stated before, increasing the shield gas flow rate results in an improved mixing process between the discharge and shield gas, which can be observed in Figures 9c,d and S2). In the case of a 3.0 slm shield gas, the interference between the main discharge gas and the additional shield gas starts to occur, although the majority of the working gas is still located closer to the surface of the table. These mixing processes lead to a higher mass fraction of argon close to the center on the surface as well as thinner argon-rich “tails,” but with a higher amount of argon in comparison to a lower shield gas flow rate. As was discussed before, a thick protective argon channel can be distinguished in the case of a 4.5 slm shield gas flow rate, which fills a significant gap volume and spreads on the surface. Therefore, by increasing the shield gas flow rate, the feasibility of the production of an assistive channel of argon is enhanced. Another eye-catching finding based on the results discussed above is the location of argon in higher concentrations using different shield gas flow rates as this location can promote plasma propagation to a remote distance. Indeed, the high velocity of shield gas due to the higher flow rate carries the remains of argon to farther distances. It can be concluded that while argon of the middle discharge gas stream only remains in a small circular area with a lower concentration when using no shield gas, the use of the shield gas assists argon from the main gas discharge stream to be transferred to farther distances across the horizontal surface.

Finally, the argon mass fraction has been compared for different shield gas types (nitrogen vs. argon), and the simulation results are presented in Figure 10 and S3. For this part of the investigation, the flow rates of the argon and nitrogen shield gases were fixed at 4.5 slm, while the distance between the additional plate and the surface was
set to 5 mm. The most significant outcome observed in Figure 10 is the potency of the argon shield gas in the formation and intensification of a channel with argon in high concentration. However, Figure 10c demonstrates that nitrogen as shield gas leads to the reduction and dispersion of argon injected from the plasma jet capillary. This means that a channel with a high concentration of argon fails to form in this case and, thus, the plasma does not have a predefined channel to propagate. Furthermore, a remarkable difference in the mass fraction of argon near the table surface for the different shield gases can be observed in Figure S3. When no shield gas is applied, argon is spread in a quite homogeneous way, while in the case of the argon shield gas, the central area contains a higher concentration of argon with three extended argon-rich "tails." At the same time, nitrogen causes shrinkage of the argon distribution resulting in only a low mass fraction of argon in a small triangle in the center of the table. All in all, it can be concluded that depending on the gas type, the shield gas can either expand or diminish the spreading of the main gas used for plasma generation, which consequently may affect the discharge in the same way. The findings of this simulation section can thus already provide preliminary insights into the footprint and dynamics of the plasma on the surface of the table.

3.3 Schlieren imaging

To thoroughly comprehend the fluid perturbation in the gap volume, flow visualization by means of Schlieren imaging was also conducted. The principle of this photography technique is based on variations in the refraction index in a medium. As such a change occurs due to the fluctuation in, for instance, gas density, this method is suitable in this investigation where different gas types are being used. Both plasma off and plasma on regimes will be visualized in this study referring to the gas stream only and the operating plasma, respectively. Figure 11 shows the Schlieren photographs for both regimes of the plasma jet using different gap sizes and fixed main and shield gas flow rates of 1.0 and 4.5 slm, respectively. In the plasma off regime, the argon flows of the main and shield gas streams encounter the quartz target, and the combined perturbed air and argon subsequently spread across the surface. On the contrary, the situation is drastically different in the presence of the discharge. When the plasma is ignited, the turbulence effect is intensified due to either gas heating, ion momentum transfers to neutral gas molecules, or a local pressure increase in the powered electrode's proximity. Later on in this study, it will be demonstrated that high-velocity plasma bullets characterize the plasma jet's structure. These plasma bullets can also induce a turbulence effect by transferring momentum between ions and neutrals.

The distance between the plasma outlet and the target is an extremely sensitive parameter influencing the flow pattern and plasma expansion. Therefore, this parameter was also varied in this study and the obtained images (shown in Figure 11) clearly demonstrate different mixing patterns of gas/plasma and ambient air. The first significant result is the extensive mixing of gases in the smallest enclosed volume (5 mm distance...
from the target) which generates an intensified turbulent flow, especially for the plasma on the regime. The enhanced contrast in Figure 11a compared to Figure 11b also validates the earlier obtained results in Figures 6a,b and 8a,b as a higher concentration of argon with a greater velocity is present in the volume between the additional plate and the surface when the distance is 5 mm. Under the two experimental conditions shown in Figure 11a,b, the excess amount of argon trapped in the comparatively small, enclosed volume overflows from the edge of the quartz plate which also supports the results obtained with CFD simulations in the previous section. Distinct and particular fluid dynamics can also be observed for the 13 mm capillary–sample distance in Figure 11c, where the volume is sufficiently large to avoid significant mixing between the discharge and shield gases and, thus, to avoid subsequent turbulence. However, the meeting point of the gases in the case of a 13 mm distance can be clearly observed on the Schlieren images for this experimental condition. The observed behavior is therefore also in line with the combination of the velocity magnitude and overall argon mass fraction distributions obtained in the CFD simulations section.

In the next step of the Schlieren imaging experiment, the investigation focuses on the impact of the shield gas flow rate on the fluids perturbation pattern between the plasma jet and the quartz table. In the absence of shield gas (Figure 12a), argon from the main gas inlet propagates in high concentration only up to an approximately 20 mm distance from the center which is in excellent agreement with the earlier obtained results (see Figure 9a). As the rest of the volume under the additional plate is uniformly filled with an argon/air mixture and the argon content is small, Schlieren images visualize this volume in a uniform color without drastic changes in contrast. On the contrary, the fluid dynamics instability and turbulence start by introducing the argon shield gas. The Schlieren images in Figure 12b–d demonstrate an increase in contrast with increasing shield gas flow rate which can be attributed to the presence of argon in higher concentration and greater gas velocities as was observed in the CFD simulations section (see Figures 7 and 9). The mentioned circumstances lead to an intensified perturbation and progressive expansion of argon which is observed as a fluid spreading over the quartz plate’s edge. Altogether, injection of the argon shield gas facilitates the presence of argon in higher concentrations in the partially closed volume, which can prompt plasma propagation. Later on in this article, the bottom view of the plasma bullets propagation will indeed clearly show that the plasma actually travels over a longer distance and spreads further when shield gas is introduced.

The influence of the shield gas type on the fluids’ perturbation behavior is further visualized with Schlieren photographs in Figure 13. The main differences in the obtained results between the conditions without any shield gas and with the presence of the argon shield gas are already discussed in the paragraph above. As such, this paragraph only focuses on the particular fluid
FIGURE 12 Schlieren images of the fluid dynamics for plasma off and on regimes obtained for a 5 mm gap size using (a) no shield gas, (b) a 1.5 slm, (c) a 3.0 slm, and (d) a 4.5 slm shield gas flow rate

FIGURE 13 Schlieren images of the fluid dynamics for plasma off and on regimes obtained for a 5 mm gap size using (a) no shield gas, (b) a 4.5 slm argon, and (c) a 4.5 slm nitrogen shield gas
behavior in the case of nitrogen shield gas. First, according to the contrast of the images, it is evident that the density in the enclosed volume is less disturbed with nitrogen injection as one of the main components of ambient air is nitrogen. Moreover, unlike argon, the nitrogen shield gas prevents the further spreading of argon/plasma from the main discharge gas stream. The latter can be explained by the unsuccessful energy transport between the reactive species and, thus, ceases the plasma expansion to farther distances. Moreover, based on the CFD simulation findings (see Figure 10), the nitrogen shield gas restricts the argon discharge gas to a small central column, as can also be seen in Figure 13c.

3.4 | OES

The spatial evolution of the reactive species optical emission intensity available in the plasma jet for different APPJ set-ups has already been investigated elsewhere. [43–45] This study, therefore, mainly focuses on the determination of the amount and type of radiative excited plasma species present in the plasma effluent propagating over the quartz surface. For this purpose, space-resolved OES measurements were performed each millimeter along the symmetry axis, making use of an optical fiber placed closely beneath the quartz plate, as indicated in Figure 1a. Distinct optical emission spectra were observed within a circle with a radius of 15 mm; thus, OES measurements were carried out from $-15$ to 15 mm with 0 mm belonging to the center of the plasma jet core. During all measurements, no new excited species were found regardless of the change in shield gas flow rate or shield gas type, which proves the inability of the shield gas in altering the type of reactive plasma species. A detailed allocation of the most intensive transition lines/bands and the corresponding wavelengths can be found in Table S1. The variations in the emission intensity of the OH rotational band $A^2\Sigma^+ \rightarrow X^2\Pi$ at 308.9 nm, the N$_2$ second positive band $C^3\Pi_u \rightarrow B^3\Pi_g$ at 337.1 nm, the atomic Ar I $2p_2 \rightarrow 1s_2$ line at 696.5 nm, and the O I $3p^3P \rightarrow 3s^5S$ line at 777.5 nm are predominant; hence, the focus of this section will be on the spatial evolution of the above-mentioned bands and lines. The OES data also reveal that the intensities of the O line and OH band follow the same trend as the Ar line and, thus, will, therefore, not be discussed in this paper. The presence of the O and OH species is believed to be due to the fragmentation of H$_2$O and O$_2$ molecules. The identified oxygen-containing molecules are present in the plasma due to the inevitable diffusion of the ambient environment and the presence of impurities in the gas-containing system. [46,47] However, as the behavior of Ar and O and OH is following the same trend it can be assumed that the main contributor to OH and O is introduced with the discharge gas meaning that the source of oxygen and OH is mainly impurities from the gas bottle. This has also been shown by analyzing the reactive species inside the capillary where traces of oxygen species were found inside the effluent before reaching the nozzle outlet. [43] Furthermore, the diffusion of ambient air into the afterglow of the plasma jet also results in the appearance of N$_2$ reactive components, which are detectable by OES. [48] In fact, the concentration of such diffused air in the plasma jet increases for the remote distances from the jet effluent core, and nitrogen molecules present in air can be easily excited by electrons or by the energy transfer from Ar excited states to N$_2$. [49,50] These electrons are probably generated due to various effects such as photoionization and a high external electric field. [31,52]

In the first step of the OES measurements, the influence of the gap size between the additional plate and the table on the intensity evolution of the acquired spectra is investigated. As previously mentioned, the measurements were carried out each 1 mm horizontal distance along the symmetry axis. The center of the plasma jet is set as zero point while the negative values represent the part of the symmetry axis under one of the used shield gas tubes (Figure 1a). Figure 14 depicts the intensity profile of the N$_2$ and Ar excited species as a function of lateral distance for different gap sizes when using a 4.5 slm argon shield gas flow rate. It is evident that the obtained distributions do not follow a symmetrical pattern which can be explained by the unique geometry of the designed plasma jet set-up. The elevated optical emission intensity of the Ar line at the negative part of the symmetry axis is due to the injected argon from the shield gas tube located above this region which acts as a source for the continuation of plasma towards the tube. However, this effect is more pronounced for the smallest gap size. These results are in excellent agreement with the simulation outcomes discussed and presented in Figure 8 where it was demonstrated that generally a higher concentration of argon near the surface volume was located on the side of the shield gas injection. As shown in Figure 14a, the intensity of the Ar line at 696.5 nm in the plasma jet center considerably increases when reducing the distance between the plasma jet and the table. From the jet center on, the Ar line intensities also rapidly decay at the more distant lateral positions. The evolution of these profiles for different capillary–substrate distances thus confirms the better mixing between the argon shield gas and the argon discharge gas at smaller distances, as was already
demonstrated in the previous sections. This effect at small distances can be validated by the confinement of the volume between the additional plate and the table as a consequence of the particular geometry of the designed plasma jet set-up. Therefore, by decreasing the gap size, it is possible to enhance the production of reactive Ar species. On the contrary, the optical emission intensities of the N$_2$ excited species follow a different pattern. Two things should be considered for understanding the trends in N$_2$ optical emission intensity. First of all, as explained before, the energy transfer between the excited atoms of Ar present in the plasma jet and the nitrogen molecules present in the ambient air leads to the production of excited states of N$_2$ species. Second, the intensity of N$_2$ optical emission directly correlates with the nitrogen concentration in the detection area. Thus, a proper balance between the available excited Ar states and nitrogen molecules is required to produce a significant amount of excited N$_2$ species, explaining the drastic difference between the OES results shown in Figure 14a, b. In other words, the product of the following two functions has a significant impact on the curve shapes observed in the second figure: (1) distribution of the Ar excited species in the region of interest and (2) distribution of nitrogen molecules at the analysis positions. The first function closely resembles the curves in Figure 14a, while the second function is more complex and directly correlates with the gas dynamics and air mixing. Evaluating the latter processes for the smallest gap size while considering earlier obtained results (Figures 8a and 11) shows that the distribution of nitrogen molecules is asymmetrical. This function has one well-defined local minimum at the center where the argon atoms from the main capillary dominate in the area and two wings at the sides with different nitrogen molecules concentrations at specific distances from the center on the left- and right-hand sides. On the left-hand side (under the shield gas tube), the nitrogen concentration is higher than on the right-hand side (between the shield gas tubes). This can be explained by the fact that ambient air and, thus, nitrogen is pulled towards the center together with the shield gas flow (left-hand side), which flows in the same direction, while on the opposite side, the stream of shield gas pushes air away from the center (right-hand side). Thus, the product of Ar excited states and nitrogen molecules distribution functions for a 5 mm gap size results in an asymmetrical curve with two maxima of different height 2–3 mm away from the local minimum value in the center, as can be observed in Figure 14b. Increasing the distance between the additional plate and the table to 10 mm results in the positioning of the N$_2$ optical emission intensity maximum in the central area around the plasma core, and this intensity rapidly declines further away from the center. As no radical changes are observed for excited Ar species distribution for a 10 mm gap size apart from the decrease in the maximum intensity (see Figure 14a), such a drastic change in the curve shape in Figure 14b can be explained by alternation in the presence of nitrogen molecules in comparison to a 5 mm gap size. Now, a considerably higher amount of nitrogen molecules diffuses towards the plasma jet center (see Figure 11b), providing species for N$_2$ optical emission. However, the obtained curve of the luminescence is not symmetrical due to the same

**FIGURE 14** Evolution of the optical emission intensity of (a) Ar (696.5 nm) and (b) N$_2$ (337.1 nm) along the symmetry axis for different distances between the additional plate and the table obtained for a 4.5 slm argon shield gas flow rate.
reasons, as described above. The left-hand side of the curve is more elevated than the right-hand side, as in the first case, the ambient air is dragged together with the shield gas flow (under the shield gas tube), while in the second case, the concentration of nitrogen molecules is reduced by the outflowing gas mixture (between the shield gas tubes). Finally, the largest distance between the plasma jet and the table leads to an overall decrease in the intensity of the optical emission spectra. Despite the high accessibility of nitrogen for this gap size compared to the other experimental conditions, the number of available excited Ar species is four and three times lower than obtained for a 5 and 10 mm gap size, respectively (see Figure 14a). Thus, as a result, the optical emission of the N₂ band at 337.1 nm for a 13 mm gap size has the lowest peak intensity and the least asymmetrical behavior.

Subsequently, the impact of the shield gas type on the intensity of the Ar and N₂ spectral line/band was investigated, and the results obtained for a 5 mm gap size and a 4.5 slm shield gas flow rate of each type along with no shield gas injection are demonstrated in Figure 15. It is worth mentioning that the curves corresponding to the nitrogen shield gas injection have symmetrical profiles regardless of the excited species that are examined. This phenomenon can be explained by the domination of argon in the small gap, which is the primary source of the excited species and, thus, prescribes the distribution of Ar and N₂ optical emission. When no shield gas is used, plasma uniformly spreads on the dielectric table, assuring symmetrical OES profiles. Adding a 4.5 slm of nitrogen shield gas does not significantly alter the profiles' symmetry as the concentration of nitrogen molecules, which can participate in energy transfer with Ar excited species, is almost the same on both sides from the center: under and between the shield gas tubes. Moreover, the deviation between these two sides is smaller than the precision of OES used in this study. On the contrary, using argon as a shield gas asymmetrically increases the number of excited Ar atoms due to a higher concentration of argon in the small volume between the additional plate and the table as was already demonstrated in Figure 10b. Injection of the nitrogen shield gas increases the optical emission intensity of the N₂ band at 337.1 nm, as can be seen in Figure 15b, which is a logical result due to the addition of nitrogen gas. Moreover, in the same figure, the lowest intensity of the N₂ line can be observed when no shield gas is added. A sharp drop in the maximum intensity value (almost four times) is observed for the plasma jet alone or when argon is used as a shield gas. For these latter experimental conditions, the optical emission profiles in Figure 15b have two smaller maximums at a radial distance of 2–3 mm from the plasma core. This peculiar behavior, as was already explained above, can be explained by the following two factors: (1) mixing processes between ambient air and the plasma jet and (2) the energy transfer balance between the excited states of Ar and nitrogen molecules. These two aspects also justify the domination of the N₂ band optical emission intensity obtained using argon shield gas at further distances from the center in comparison to the other experimental conditions as in this case, the mentioned processes occur at even higher rates leading to an increased number of excited nitrogen molecules. In addition, as mentioned before, argon shield gas fills more

![Figure 15](image-url)
volume in the central area compared to no shield gas and thereby limits the diffusion of air in this region resulting in the lowest intensity of the N2 line in the central region. This observation clearly reflects the capability of the shield gases in the excitation of N2 molecules through different patterns.

Additionally, this study also examines the impact of different gas flow rates of nitrogen and argon shield gases on the intensity profiles of the Ar and N2 optical emission for a fixed 5 mm distance between the plasma jet outlet and the table and the results are demonstrated in Figure S4. The profiles of the emission intensity of the Ar line follow the same trend as in Figure 15a. This observation is in good agreement with the simulation and Schlieren imaging results shown in Figures 9 and 12 where the exhibition of mass fraction and fluid perturbation for different argon gas flow rates suggested that with the highest shield gas flow rate the main argon stream merges with the injected shield gas. This joined flow fills the entire volume beneath the additional PVC plate suggesting a better development of the plasma in this region. When using nitrogen shield gas at different gas flow rates, all Ar line profiles overlap proving that the nitrogen gas flow rate does not affect the optical emission intensity of the Ar line, as demonstrated in Figure S4c. Indeed, the argon intensity cannot increase as it would require extra argon atoms or additional power; thus, it remains the same as observed for the plasma jet without any shield gas. Moreover, there is another pathway to obtain photoemission of N2, which involves the direct excitation of nitrogen with electrons.[53] Therefore, adding extra nitrogen shield gas to a small gap will not significantly influence the intensity of the Ar optical emission. A higher flow rate of any of the used shield gases also promotes the excitation of nitrogen molecules, thereby resulting in an increased intensity of the corresponding optical emission, especially in the case of nitrogen shield gas (Figure S4b,d). The higher flow rate of argon shield gas pulls more air towards the plasma jet resulting in an increase of nitrogen molecules concentration, especially on the left-hand side of the plasma jet, under the shield gas tube. Furthermore, at the same location, a pure source of argon shield gas flowing out of the tubes is injected towards the plasma effluent creating a continuation to plasma and generating higher concentrations of reactive species in that pathway. The explanation mentioned in the previous paragraph about the gas mixing and N2 excitation processes can also justify the results shown in Figure S4b,d.

3.5 | ICCD imaging

At first sight, an AC plasma jet seems to be a continuous discharge; however, it is well-known by now that it actually consists of a series of bullets propagating with high speed.[54,55] As soon as the bullets impinge a surface, they spread on the surface, which makes it practical to perceive how far their pattern can spread on a particular substrate.[56] Such information is beneficial to understand the propagation of the plasma jet on flat surfaces as well as to explain other observations. Therefore, the APPJ footprint on a dielectric surface was visualized through high-speed imaging by using an ICCD camera. The camera was adjusted under the quartz window to gather images of the propagation of plasma on the transparent surface from the bottom view, as demonstrated in Figure 1a. Figure 16 shows the evolution of the plasma footprint on the surface of the quartz window for a 5 mm distance between the capillary edge and the surface without the injection of shield gas. On the basis of the OES spectra acquired and analyzed in the previous section, it can be concluded that the primary sources for light emission are excited Ar and N2 species.

Figure 16: Plasma footprint on the surface of the quartz window obtained for a 5 mm distance between the additional plate and the surface without shield gas over 90 ns time
Therefore, it can be expected that the main visible channel in the proximity of the plasma jet core mainly consists of excited Ar atoms which are followed by the diffusion of N$_2$ molecules from the surrounding air and the subsequent excitation of N$_2$ through interaction with plasma.

The photos in Figure 16 illustrate the evolution of the plasma bullet spreading patterns on the surface over 90 ns. On the basis of the first photo (Figure 16a), primarily, the bullets collide with the quartz plate initiating their propagation from the central point. Moreover, at this moment, a bright branched structure is clearly visible due to the high concentration of the excited species that were generated inside the plasma jet core and that reached the surface. Afterward, the excited species travel further on the surface and spread across a more substantial area (Figure 16b,c), which in turn leads to a decrease in the concentration of the excited species. This factor, together with the plasma decaying and its mixing with air, causes the reduction in the registered pattern brightness. This latter observation is in excellent agreement with the OES results discussed above.

Figure 17 reveals the development of the plasma discharge structure on the surface for a 4.5 slm argon shield gas flow rate using different gap sizes. The first photo from this figure demonstrates that for a 5 mm distance between the plasma jet capillary and the quartz window, the branches are highly elongated and they spread farther from the effluent center. Indeed, based on the findings from Figure 14, a higher number of excited plasma species was observed when using the shortest distance between the additional plate and the surface. Moreover, the simulation outcomes shown in Figure 8 and the gas flow visualization through Schlieren imaging in Figure 11, demonstrate the development of a channel with a higher argon concentration for a 5 mm gap size which suggests that the plasma can propagate through this channel to further distances. These two aspects explain the drastic difference between the ICCD images obtained for the shortest plasma-surface distance and the other distances under investigation. By increasing the distance between the plasma jet capillary and the quartz window, the length of the plasma footprint pattern is strongly attenuated. This observation can be explained by the decrease in the argon mass fraction on the surface obtained from simulations as well as the lower intensity of the Ar line in the OES spectra (Figures 8 and 14a, respectively).

Finally, the influence of the shield gas type and its flow rate was examined by looking at the distance the plasma footprint can propagate on the dielectric surface without detaching from the plasma jet core. As seen in Figure 18, a considerable dependency of the branches’ length on the gas flow rate can be observed, especially for the argon shield gas, while this influence is less pronounced in the case of nitrogen shield gas. On the basis of the shown ICCD images, using argon as shield gas with a flow rate of 4.5 slm can stimulate the propagation of the plasma footprint up to a 30 mm radial distance from the center of the jet. This observation is in accordance with the simulation results in Figure 9, where a higher concentration of argon atoms close to the surface is obtained for a higher flow rate of argon shield gas. However, when nitrogen is used as a shield gas, no significant differences were observed on the ICCD images in terms of plasma footprint spreading. As was demonstrated by the simulation results in Figure 10c, this type of gas leads to a reduction of argon atoms and their excited states close to the surface due to mixing and energy transfer processes. Thus, injection of nitrogen to the shield gas system reduces the area of the plasma jet footprint spreading which means that nitrogen gas acts as a hindrance against further propagation of the head of the plasma jet bullets. Furthermore, the fluid dynamic visualization in Figure 13 clearly depicts the reduced propagation of plasma when introducing nitrogen shield.
gas. The evidence to this conclusion can be observed on the ICCD images shown in Figure 18 when comparing the photo of only the plasma jet (no shield gas) and any of the photos taken of the plasma jet with nitrogen shield gas. The OES results presented in Figure 15 and S4 provide a complementary explanation of the results observed in Figure 18 as the intensity of the optical emission observed in this study can be ranked in the following order: “argon shield gas” > “no shield gas” > “nitrogen shield gas.”

Another ICCD imaging set-up with the camera placed perpendicular to the plasma jet axis was used to visualize the spatial and temporal development of the plasma plume and, thus, to obtain more detailed insights on the discharge dynamics. The timescale at which the plasma evolves is in the order of nanoseconds which is relatively short to observe changes in detail. However, the repetition of the plasma evolution is temporally stable so averaging over multiple periods acquires the desired information with a sufficiently high temporal resolution. The obtained results of this ICCD imaging technique are consistent with the findings of the previous imaging set-up where the camera was located underneath the table, perpendicular to the quartz substrate. Figure 19 presents the series of ICCD images of the time-resolved evolution of plasma jet propagation on the surface for a 5 mm capillary–sample distance during the positive and negative half-cycle. On the basis of this figure, the positive half-cycle is responsible for visual plasma propagation on the surface, while during the negative half-cycle, the plasma appears as a thicker and more confined discharge in the central region. Therefore, the discussion will be focused on the positive half-cycle. In the series of images on the left column of the figure, it is observable that in the beginning, the plasma plume starts to flow outside the capillary, and, at the target, the axial movement is turned to a radial spread along the glass surface. At this phase, as the quartz target hinders the plasma bullets, the charges accumulate on the surface. From here, the discharge propagates on the surface in a radial direction. If the jet propagates significantly farther, the connection of the plasma to the initial point (plasma bullet afterglow

**FIGURE 18** Plasma footprint on the surface of the quartz window for different shield gas types and flow rates

**FIGURE 19** Discharge development in time for positive and negative currents with a 5 mm distance between the additional plate and the quartz window surface without a shield gas
(tail) starts to fade away. In the last phase, after 1.4 µs, the plasma is fully spread on the surface, and the plasma branches reach their maximum lateral distance on the quartz surface, where simultaneously the connection to the jet outlet is almost fully quenched. This can be explained as the excited argon states outside the tube are in direct contact with air, which means the presence of nitrogen and oxygen species, and consequently, the quenching process starts to dominate. This observation is in line with Figure 16c, where the dissipation of a bullet in the central region is demonstrated. The same explanation as was used for that image is valid here as well: the transferred energy is consumed to propagate plasma to further distances while at the same time, the reactive species are mixed with the surrounding air; thus, the plasma bullets do not propagate further.

For the sake of easy comparison between different experimental conditions, all further discussion will focus only on the images where the plasma is completely propagating on the surface. For the temporal discharge evolution on the quartz surface, videos of the collected sequential photos can be found in the Supporting Information.

The dependency of discharge propagation on different gap sizes with a 4.5 slm argon shield gas is demonstrated in Figure 20. It is evident that the propagation pattern is different for each gap size variation as the propagation changes from the moment the bullet collides with the surface. First, in a 5-mm capillary–table distance, the plasma travels substantially to further lateral distances as a high concentration of argon enables the plasma to propagate remoter, whereas, for the 10 and 13 mm gap sizes, the air mixing with plasma ceases the propagation process as explained earlier with CFD simulations and OES findings. Another interesting outcome observed in Figure 20 is that the plasma is still connected to the jet outlet for gap distances above 5 mm. Likewise, in Figure 17, it can be observed that the plasma core is brighter for larger gap sizes. In fact, plasma most likely will follow the path where the highest concentration of working gas is available. In this study, two sources supply argon for plasma propagation: the main argon channel and the argon shield gas. As demonstrated in Figure 8, such an argon-rich region is mainly located in the sample center for higher distances. In the case of a 5 mm gap, a weak connection between the plasma bullet expanding on the surface and its source of origin might be due to the intensified turbulent flow in the center or a charge accumulation in the spreading area.

Variation in argon shield gas flow rate with a fixed 5 mm distance between the capillary and the quartz window results in different plasma bullet spreading patterns, as was observed with ICCD imaging, and is presented in Figure 21. Those captured discharge developments reflect the combination of a few phenomena. First of all, different gas dynamic regimes in each case were already demonstrated in this study (Figures 7, 9, and 12). Second, depending on the shield gas flow rate, the plasma bullet contains a different concentration of reactive species (Figure S4), which will result in changes in radiation detected by the ICCD camera. A combination of these processes results in different plasma bullet spreading patterns on the quartz surface and the presence of the connection to the plasma.

**Figure 20** Discharge development on the quartz window surface obtained for a 4.5 slm argon shield gas flow rate and different gap sizes

**Figure 21** Discharge development on the quartz window surface obtained for a 5 mm gap size and different argon shield gas flow rates
jet capillary, as, for instance, plasma discontinuation from the jet outlet is visible when the plasma is fully spread on the surface using 3.0 and 4.5 slm of shield gas flow rates.

Finally, the impact of shield gas type on the plasma jet dynamics has been investigated with ICCD imaging in the perpendicular direction. The obtained results for a fixed 5 mm gap size and a 4.5 slm shield gas flow rate of nitrogen and argon are presented in Figure 22. The aspect of plasma bullet propagation in the case of no shield gas or argon shield gas has already been discussed above. The most interesting outcome of Figure 22 is the distinctive plasma propagation pattern while using nitrogen shield gas. The plasma brightness in the central area represents the higher emission intensity in the core and the higher charge accumulation in that particular region when nitrogen is used as the shield gas. This luminosity in the central area of the target for the same experimental condition was further observed in Figure 18. Moreover, in the CFD simulation section of this study (Figure S3 and 10), nitrogen shield gas has already revealed the potency to constrain the argon distribution upon the target. Such working gas confinement results in the brighter plasma emission in the substrate central region and nicely shows the harmony between all obtained results.

4 | CONCLUSION

In summary, this study intended to evaluate the potency of using a shield gas to control and manipulate APPJ properties and its composition to achieve a desired well-defined environment. First, variations in discharge power for different distances from the capillary to the table were calculated using $I-V$ waveform curves. The obtained results indicate that a higher flow rate of Ar shield gas results in a considerable increase in discharge power, especially with a 5 mm gap size and a 4.5 slm argon shield gas flow rate. In contrast, using a nitrogen shield gas results in no significant difference in the calculated discharge power. Second, CFD simulations provide details regarding the gas mixing processes and the mass fraction distribution of argon in the volume between the additional plate and the table. The major finding is the creation of a region with high concentrations of argon when argon gas was additionally injected while the use of nitrogen as shield gas prevents the formation of such a region. Complementary to the simulation results, fluid dynamics visualization using Schlieren imaging shows the high perturbation of gas in a smaller gap size and a higher flow rate of argon shield gas. In contrast to argon, injecting nitrogen into the plasma system limits the volume of the highly concentrated argon and reduces fluid mixing. The performed OES measurements of the most intensive lines and bands reveal an asymmetric behavior in the case of argon shield gas, which is explained by the location of the shield gas inlets. The highest intensity of the optical emission Ar line is observed for the shortest gap and decreases when using larger gaps. The distinctive pattern of the $N_2$ band emission intensity observed for the shortest distance is explained by the shielding effect of injected argon, the mixing with ambient air, and the energy transfer processes from the excited states of Ar to nitrogen molecules. Additionally, it is demonstrated that the shield gas flow rate is an influential parameter in the alteration of the intensities of the reactive species optical emission especially at larger distances from the plasma jet center, as a higher flow rate of argon shield gas enhances the intensity of the Ar line. Finally, high-speed ICCD imaging with two different configurations reveals a significant dependency of the plasma footprint propagation distance on the applied experimental conditions. Increasing the gas flow rate of argon shield gas leads to long plasma branches due to the excess value of excited species present at distances farther away from the jet core. In contrast, the introduction of nitrogen gas causes shrinkage of the plasma footprint pattern on the surface and thus the higher luminosity of the plasma jet upon collision on the surface. Overall, the findings of this study provide valuable insights into the properties of an atmospheric pressure plasma jet with different shield gases and various working parameters with an excellent agreement between the outcomes from various techniques. The shield gas type and its flow rate are found to have the most significant impact on the plasma properties which might be useful information in future work on the treatment of polymeric surfaces, effective deposition, and the exclusion of unwanted species during the utilization of APPJs for biomedical applications.
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CONFLICTS OF INTEREST
The authors declare no conflicts of interest.

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