

# Influence of helium mole fraction distribution on the properties of cold atmospheric pressure helium plasma jets

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## ABSTRACT

The influence of helium mole fraction distribution in air on the cold atmospheric plasma jets excited by 1.5 kHz rectangular high voltage pulse is studied in this work. Computational fluid dynamics (CFD) with incorporation of large eddy simulation (LES) model is used to simulate the helium mole fraction distribution in air under the helium flow from laminar to turbulent regime with increasing helium outlet velocity. Numerical simulation results are combined with experimental results in order to determine the influence of helium distribution on the cold plasma jets. It reveals that the structure of the helium distribution caused by diffusion or by turbulent mixing in turbulent regime determines the characteristics of the cold plasma jets. On the other hand, the curves of plasma jet length ( $L$ ) versus helium outlet velocity ( $V$ ) at different jet diameters ( $D$ ) are unified in a map of jet Reynolds number ( $Re = \rho_{He} \cdot V \cdot D / \mu_{He}$ , where  $\mu_{He}$  is the helium viscosity constant) versus dimensionless plasma jet length ( $l = L/D$ ). The map is allowed to predict the flow pattern of helium jet in order to estimate and control the plasma jet length at different jet diameters.

## I. INTRODUCTION

Cold atmospheric plasma jets recently have received a lot of attention in view of many possible applications such as materials processing<sup>1-3</sup>, biomedical surface modification<sup>4-6</sup> and synthesis of nanomaterials<sup>7-9</sup>. Recent numerical simulations and experimental studies have made significant advances in the understanding of the cold plasma jets<sup>10-16</sup>. The detailed studies have revealed that the cold plasma jet produced by nano-second or micro-second pulsed discharges is typically composed of a series of rapidly propagation streamer discharges. The streamer with a luminous head propagates at very high speed in order of  $10^4$ - $10^5$  m/s over several centimeters<sup>15, 17, 18</sup>. The experimental images reveal that the luminous zone of the helium plasma jet is a ring-shaped [13], and the peak of luminance is shifted off the jet axis<sup>19</sup>. Since  $N_2$  and  $O_2$  have a higher breakdown voltage than helium, the peak ionization occurs in the He-air mixture zone where sufficient helium neutrals are present and the electric field magnitudes are high enough<sup>12</sup>. Therefore, the distribution of helium in air defines the spatial region where the reaction between helium and air such as the penning ionization reaction ( $He^m + N_2 \rightarrow N_2^+ + He + e$ ) can occur. Previous studies have emphasized the importance of this reaction in the cold helium plasma jet<sup>16, 20</sup>. On the other hand, the helium distribution in air also has a significant effect on the streamer propagation length<sup>20-22</sup>. Reference<sup>20</sup> reveals that a minimum helium mole fraction is necessary to sustain the plasma streamer propagation under different helium outlet velocity, and the reference<sup>21</sup> proves that length of streamer propagation in pure helium is much longer than in the helium-air mixture. All of the results demonstrate that the helium distribution in air is considerably important for the helium cold plasma jet. However, the effect of the helium distribution in air on the cold plasma jet is still not fully understood. In particular, the influence of the helium flow under turbulent flow regime on the plasma jet is unclear.

For this study, we use a computational fluid dynamics (CFD) with incorporation of large eddy simulation (LES) model to simulate the helium mole fraction distribution in air under laminar and turbulent flow regime. We combine the results obtained from the numerical simulation and the experiment in order to determine the influence of helium mole fraction distribution on the cold plasma jet for specified pulse discharge. In addition, we present the length of plasma jet ( $L$ ) as a function of outlet helium velocity ( $V$ ) at different jet diameters ( $D$ ). Also, the  $V$ - $L$  curves for different  $D$  are unified in a map of  $Re$  versus  $l$ , where  $l$  ( $l=L/D$ ) is the dimensionless plasma jet

length) and  $Re$  is jet Reynolds number.  $Re-l$  map has a value of application. Under a constant of applied voltage pulse, only one of  $V$  versus  $L$  graphs at a certain jet diameter is completed and the other  $V-L$  will be calculated at different jet diameters. On the other hand, we can use the Reynolds number to predict the flow pattern of helium jet flow and estimate the plasma jet length at different jet diameters in this flow pattern.

## II. EXPERIMENTAL SET-UP AND CFD MODEL

The scheme of the experimental setup is shown in figure 1. The setup used in this work was reported in our previous investigation on the argon plasma plume<sup>23</sup> The setup is composed of a plasma jet device, a HV pulse power supply, a gas flow system and a measurement system including an ICCD camera and an oscilloscope. The HV electrode inserted into the syringe quartz tube is arranged inside and aligned along the axis of the tube. The syringe tube can be changed in order to adjust the diameter of jet outlet where the helium flow ejects into the surrounding air. The amplitude of the applied voltage pulse is 8 kV with 5  $\mu$ s pulse width and a frequency of 1.5 kHz. The helium flow rate is measured and controlled by using the mass flow controller in the range of 50 SCCM to 50 SLM. The applied voltages is measured by a Tektronix P6015 A 1000:1 high-voltage probe and the current by an IPG current transformer. The photographs of the helium plasma jet were obtained by using a Hamamatsu ICCD camera (C8484) at fixed exposure time of 50 ms.

Computational fluid dynamic simulation for the helium mole fraction distribution is performed by the commercial CFD software code ANSYS Fluent (ANSYS Fluent 12.1). The code solves the basic conservation of mass and momentum equations. A description of the models, examples and related articles may be found in the user's guide and tutorial guide<sup>33</sup>. The code contains the LES model which is used to simulate the turbulence of the helium jet flow when the helium outlet velocity is high enough. More details about LES model can be found in reference<sup>24</sup>. The governing equations of the LES simulation are as follows<sup>25</sup>:

Conservation of mass and transport of individual gaseous species,

$$\frac{\partial}{\partial t}(\rho \cdot Y_i) + \nabla(\rho Y_i \vec{u}) = \nabla(\rho D_i \nabla Y_i) \quad (1)$$

Where  $Y_i$  is the mole fraction of the individual gaseous species;

Conservation of continuity,

$$\frac{\partial}{\partial t}(\rho) + \nabla(\rho\bar{u}) = 0 \quad (2)$$

Conservation of momentum equation:

$$\frac{\partial}{\partial t}(\rho\bar{u}) + \nabla(\rho\bar{u} \cdot \bar{u}) + \nabla p = \rho\bar{g} + \bar{F} + \nabla \cdot \tau_{ij} \quad (3)$$

LES computes directly the large-scale eddies, and the sub-grid scale dissipative process is modeled by using sub-grid models (SGM). The dynamic kinetic energy subgrid-scale model is chosen as the SGM model in this work. More detail about this SGM model can be found in the references<sup>26-28</sup>. The size of computational domain in this simulation is shown in figure 2 (a). A domain of 100 mm wide, 100 mm long and 200 mm high is built. In the LES simulation, the grid size is an important factor to be considered. The grid should be fine enough to include the turbulence scales associated with the large eddy motions that can be resolved accurately enough by SGM. The balance should be considered for the grid size and the computation ability. The mesh in the axial plane used in the simulation is shown in figure 2 (b). The largest grid size is 5X5X5 mm<sup>3</sup> and the smallest one is (5/16)X(5/16)X(5/16) mm<sup>3</sup> (total 308208 cells). The inlet velocity is set as the inlet boundary condition as shown in figure 2 (a) and the remaining boundaries are specified as pressure outlet boundary.

### III. RESULTS AND DISCUSSION

#### A. The effect of helium distribution in laminar flow

Figure 3 displays the photographs of the plasma propagation sustained at different helium flow rate. The plasma jet length increases almost linearly with the helium flow rate, when the helium flow is laminar. The same result is also obtained at other different diameters of jet as shown later. As discussed earlier, the helium-air diffusion zone is essential for the propagation of this plasma jet, and the amount of helium mole fraction should always be maintained high enough to sustain the plasma in the ionization channel. This explains why the length of helium jet core, with a mole fraction which sustains the plasma propagation, increases with the helium outlet velocity during laminar regime. The white lines in figure 1 are the distribution of the contours of the helium mole fraction in axial plane obtained from the numerical simulation. The distance in axial direction from the jet outlet to the location where it has the minimum helium mole fraction needed to sustain the plasma is extended from 23.4 mm at an outlet helium velocity of 0.85 m/s to 62.5 mm at the velocity of 2.55 m/s as shown in figure 3. To clearly reveal the role of the diffusion of

helium into the surrounding air along the axial direction at different helium outlet velocity during laminar flow, we show the helium mole fraction decay along the centreline in variable velocities in figure 4. It is obvious that the centreline decay rate is significantly higher at low helium outlet velocity, by comparison with the high helium outlet velocity. It is interesting to note that the minimum helium mole fraction which is necessary to sustain the plasma propagation is almost constant between 0.45 and 0.5 for different helium outlet velocities. However, the minimum helium mole fraction value increases with an increase in the helium outlet velocity during helium laminar flow in reference <sup>20</sup>. This may be due to the difference in applied high voltage, pulsed width and frequency used by the authors compared to this work.

For a better understanding of the effect of the helium mole fraction distribution on the plasma jet during the laminar regime, the helium mole fraction distributing in the cross-sectional plane at different axial heights is displayed in figure 5 for the helium outlet velocity of 1.70 m/s. One can clearly notice that the overall structures of helium mole fraction distribution at variable heights have completely symmetric geometry. Therefore, it verifies that the simulation for the plasma jet in <sup>11, 12, 20</sup> can be used as a two-dimensional axis-symmetric model during helium flow in laminar, while the symmetric structure disappears when the helium flow comes into the turbulent regime as shown later. Meanwhile, one can also find that more and more helium in the jet core diffuses into the stagnant ambient air as the jet develops, which leads to the decrement of the plasma jet diameter with increasing the axial height as shown by the red circles in figure 5. However, the minimum helium mole fraction to sustain the propagation in the radial direction decreases with an increase in the axial height as presented in figure 6. This indicates that the self-induced electric field <sup>12</sup> at the edge of plasma becomes stronger for increasing axial height.

## **B. The effect of helium distribution in turbulent flow**

By increasing the helium velocity, the helium flow will develop from the laminar regime to the turbulent regime. The length of plasma jet significantly decreases at the transition of the flow to the turbulent regime as describe later in this work and in references <sup>20-22</sup>. As shown later and in <sup>20</sup>, the maximum length of the plasma jet is achieved during the flow at the transition from the lamina to the turbulent regime. Articles on the effect of helium distribution under turbulent flow on the plasma jet are considerably rare. Therefore, it is necessary to investigate the plasma jet which is affected by the helium distribution caused by turbulent flow. We will display and discuss the

influence of the helium mole fraction distribution in three-dimensional with time evolution on the plasma jet during the turbulent regime in this section.

Figure 7 (a) displays the typical plasma jets during the transition from the laminar flow to the turbulence at the helium outlet velocity of 8.47 m/s and a jet diameter of 5 mm. From figure 7 (a), one can clearly observe that the plasma zone approaching to the jet outlet ( $Y < 55$  mm) is very stable, while the plasma becomes unstable when the distance is away from the exit more than 55 mm as shown in the rectangle of figure 7 (a).

To examine the role of the influence of the helium mole fraction distribution on the plasma jet under the turbulent regime, the turbulent mixing between helium and air is simulated by LES model. From the simulation results as shown in figure 7 (b), (c) and (d), the helium distribution close to the jet outlet, which is mainly caused by the diffusion, is steady and axis-symmetry as the same as in laminar flow. However, the helium distribution in the zone which is far from the outlet is unsteady and has an asymmetric structure caused by the turbulent mixing of helium and air. One can observe that the helium jet core is disturbed by the perturbation along the helium jet core causing the instability of the core. The vortex which develops from the instability helium jet core causes to the turbulence mixing helium with air. Therefore, the helium distribution in this unsteady region causes the instability of the plasma. It is important to note that the location where the instability of plasma appears to occur agrees well with the transition point where the helium jet core begins to become turbulent flow. This reveals that the characteristic structure of the helium distribution caused by diffusion or turbulent mixing determines the stability of the plasma jet. To quantify the transition between these two distinct zones of helium distribution, figure 8 shows the helium mole fraction along the centreline with time evolution. The transition point from steady flow to unsteady flow is located approximately at  $Y=60$  mm, which is good agreement with the location approximately  $Y=55$  mm where the plasma becomes instability.

To more clearly depict the distinctive feature of the mixing helium-air in these two zones, we present the helium mole fraction distribution in the cross-sectional plane at different axial heights, as shown in figure 9. It is obvious that the helium distribution is very symmetric when the cross-section is at the height of 20 mm as shown in figure 9 (a). So, the plasma is stable at this location as shown in figure 8. With the height being increased to 50 mm, the helium distribution outside jet core with low helium mole fraction appears to be asymmetric in structure, while the jet

core with relatively high helium mole fraction sustains symmetric and steady as presented in figure 7 (b). As discussed previously, the plasma always propagates along the helium jet core with high enough helium, so the helium distribution outside jet core has little effect on the plasma jet. Therefore, the plasma jet can stay stable in this case. However, the helium distribution is completely asymmetric in jet core, when the axial height achieves 80 mm. Therefore, the plasma propagating in this helium distribution is unsteady as shown in figure 8 from the experimental result.

When the helium outlet velocity is high enough, the helium flow will be transferred to the completely turbulent regime. The plasma propagation in this regime is shown in figure 10 (a). The length of plasma obviously appears to be short and the length of turbulent zone seems to be equal to the length of steady zone. The plasma length remains almost constant and is independent on the outlet velocity as shown later. We present the helium mole fraction distribution in the axial plane in figure 10 (b), (c) and (d) to explain this phenomenon. One can observe that the perturbation along the helium jet core is significantly stronger than in the transitional regime as shown previously, and the wavelength of perturbation appears to be short. Both of them give rise to the transition point, where the transition from steady flow to turbulence occurs, to approach the jet outlet as shown in figure 11. However, the intensity of the perturbation does not increase without limit as the outlet velocity increases. This limited is imposed by viscous drag and by the accelerating stagnant ambient air<sup>28</sup>. Therefore, it causes the length of the plasma jet to be short and remain constant during this regime.

### **C. The plasma jet length**

We discussed the influence of the helium distribution on the plasma jet for different outlet velocities at the jet diameter of 5 mm in previous section. As presented in figure 12 (c), one can clearly observe that the plasma length increase nearly linear with the helium outlet velocity under the laminar flow. With the outlet velocity increasing further, the helium flow reaches the transition from the laminar to the turbulent flow. Meanwhile, the plasma length goes through a maximum (~150 mm), after which, with further increase in the helium outlet velocity, starts to significantly decrease. When the outlet velocity is high enough, the plasma length nearly maintains a constant value due to the helium flow in the completely turbulent regime. A similar result was also obtained in the references<sup>28,29</sup>, but without detailed explanation that the helium distribution in the turbulent

flow causes the plasma jet to shorten. On the other hand, we also present the plasma jet length as a function of helium outlet velocity at different jet diameters as shown in figure 12 (a) and (b). The similar relation between plasma jet length and outlet velocity is obtained at the diameter of 0.6 mm and 1.8 mm. As analyzed earlier, the plasma propagation is determined by the helium distribution which strongly depends on the helium jet flow pattern at a constant of applied voltage pulse. From earlier investigation on helium jet<sup>30-32</sup>, the helium jet flow pattern can be determined by relevant dimensionless numbers, which are the jet Reynolds number (Re) and Richardson number (Ri), defined as follows:

$$\text{Re} = \frac{\rho_{\text{He}} \cdot V \cdot D}{\mu_{\text{He}}} \quad (4)$$

$$\text{Ri} = \frac{g \cdot D \cdot (\rho_{\text{air}} - \rho_{\text{He}})}{V^2 \cdot \rho_{\text{He}}} \quad (5)$$

Where  $\rho_{\text{He}}$  and  $\rho_{\text{air}}$  are, respectively, the helium and air densities,  $V$  is the helium average outlet velocity,  $D$  is the jet diameter;  $\mu_{\text{He}}$  is the helium viscosity constant and  $g$  is the gravitational acceleration. The Reynolds number gives a measure of the ratio of internal forces to viscous forces; The Richardson number is a measurement of the relative strengths of internal and buoyancy. However, when the momentum is dominated in the helium jet at Ri of the order of  $10^{-3}$ , the buoyancy is considered negligible<sup>31-33</sup>. Most magnitudes of the Richardson number in this work are of the order of  $10^{-3}$  or below, especially for the small jet diameters. Therefore, we summarized the dimensionless plasma jet length ( $l=L/D$ ) as a function of Reynolds number at different of jet diameters as shown in the figure 13. One can clearly find that when Re is smaller than 200, the helium flow pattern is under the laminar flow and the  $l$  linearly increases with the Re. When the Re is between 200 and 650,  $l$  achieves the maximum value of approximately 30 at the Re in the range from 200 to 300 and then rapidly decreases. Finally,  $l$  almost keeps a constant value of 10 due to the helium flow reaching the completely turbulent flow regime when Re is larger than 650. The Re- $l$  curve is in good approximation independent on the jet diameters. Therefore, the relationship between  $V$  and  $L$  can be calculated for other jet diameters from one L-V curve at a given discharge pulse. On the other hand, we can use the Reynolds number to predict the flow pattern of helium jet flow and estimate the plasma jet length in this flow pattern.



## IV. Conclusion

In summary, we have performed the numerical simulation of the helium mole distribution in air during the helium flow in laminar or turbulent regime, and we combined the results obtained from simulation and experiment to reveal the influence of helium distribution on the cold atmospheric plasma jets. The results indicate that the helium distribution in laminar regime, which is caused by helium diffusion, is steady and axis-symmetry. The plasma jet which is determined by the helium distribution is stability and symmetry. The length of plasma jet linearly increases with helium outlet velocity in laminar regime. In turbulent regime, the helium distribution which is caused by the turbulent mixing is unsteady and results in the instability of the plasma jet. Moreover, the results reveal that the transition point from laminar flow to the turbulence flow determines the length of plasma jet in the turbulent regime. On the other hand, the V-L curves at different D can be unified in a map of l in function of Re at a rectangular HV pulse with constant amplitude, width and frequency. Under the applied voltage pulse in this work, the helium flow pattern is under the laminar regime and the l linearly increases with the Re when Re is smaller than 200. Re in the range from 200 to 300, l achieves the maximum value of approximately 30 and then rapidly decreases. Finally, l almost remains a constant value around 10 when Re is larger than 650.

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## Captions

**Figure 1.** Experimental set up used to generate helium plasma jet propagation in helium-air mixture by pulsed voltage of microsecond width.

**Figure 2.** Schematic of the computational domain (a) and the mesh in the axial plane (b).

**Figure 3.** Photographs of the plasma jet at a helium flow rate of 1SLM (a), 2 SLM (b), and 3 SLM (c) with a jet diameter of 5 mm for 8 kV applied voltage, 5  $\mu$ s pulsed width and 1.5 kHz frequency and the helium distribution in the axial plane at the helium outlet velocity of 0.85 m/s (a), 1.70 m/s (b) and 2.55 m/s (c). The white lines are the contour of helium mole fraction obtained by numerical simulation.

**Figure 4.** Centerline helium mole fraction decay at variable helium outlet velocity of 0.85 m/s, 1.70 m/s and 2.55 m/s, corresponding to a helium flow rate of respective 1 SLM, 2 SLM and 3 SLM. A, B and C are the locations of the tip of plasma jet in respective figure 1 (a), (b) and (c).

**Figure 5.** Helium mole fraction distribution in the cross-sectional plane at different axial heights of  $Y=10$  mm (a),  $Y=20$  mm (b) and  $Y=40$  mm (c). The red lines in (a), (b) and (c) are the interfaces between plasma and air.

**Figure 6.** Radial profiles of helium mole fraction at different axial heights of  $Y=10$  mm,  $Y=20$  mm and  $Y=40$  mm;  $d_1$ ,  $d_2$  and  $d_3$  are the diameters of the plasma at  $Y=10$  mm,  $Y=20$  mm and  $Y=40$  mm cross-sectional planes in figure 5, respectively.

**Figure 7.** Photograph of the plasma jet under turbulent flow at the helium flow rate of 10 SLM with a jet diameter of 5 mm, corresponding to a helium outlet velocity of 8.47 m/s (a). And the helium mole fraction distribution in the axial plane with time evolution of  $t=135$  ms (a),  $t=195$  ms (b) and  $t=240$  ms (c). At  $t=0$  ms helium starts to come out of the jet outlet.

**Figure 8.** The helium mole fraction distribution along the centerline for a helium outlet velocity of 8.47 m/s with time evolution.

**Figure 9.** The helium mole fraction distribution in the cross-sectional plane in transition regime with different axial heights of  $Y=20$  mm,  $Y=50$  mm and  $Y=80$  mm at the moment  $t=195$  ms.

**Figure 10.** The photograph of the plasma jet in completely turbulent regime for a helium flow rate of 20 SLM with a jet diameter of 5 mm, corresponding to a helium outlet velocity of 16.94 m/s (a). And the helium mole fraction distribution in the axial plane at  $t=63$  ms (a),  $t=66$  ms (b) and  $t=69$  ms (c). At  $t=0$  ms helium starts to come out of the jet outlet.

**Figure 11.** The helium mole fraction distribution along the centerline at the helium outlet velocity of 16.94 m/s with time evolution.

**Figure 12.** Plasma jet length ( $L$ ) versus helium average velocity ( $V$ ) for variable jet diameters of 0.6 mm, 1.8 mm, and 5.0 mm for a constant 8 kV applied voltage, 5  $\mu$ s pulsed width and 1.5 kHz frequency.

**Figure 13.** Dimensionless plasma jet length ( $l$ ) as a function of jet Reynolds number ( $Re$ ) for different jet diameters.