Arc-Free Atmospheric Pressure Cold Plasma Jets: A Review

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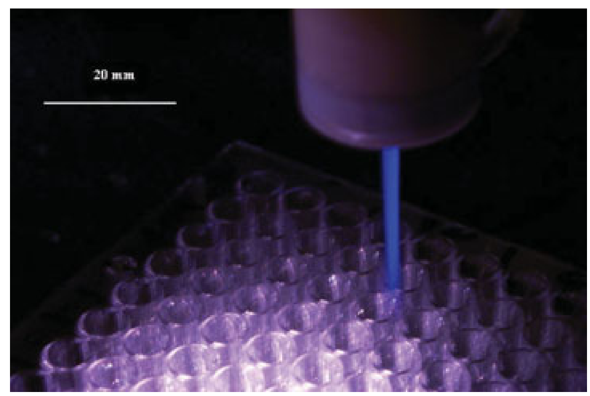
Non-thermal atmospheric pressure plasma jets/plumes are playing an increasingly important role in various plasma processing applications. This is because of their practical capability to provide plasmas that are not spatially bound or confined by electrodes. This capability is very desirable in many situations such as in biomedical applications. Various types of 'cold' plasma jets have, therefore, been developed to better suit specific uses. In this paper a review of the different cold plasma jets developed to date is presented. The jets are classified according to their power sources, which cover a wide frequency spectrum from DC to microwaves. Each jet is characterized by providing its operational parameters such as its electrodes system, plasma temperature, jet/plume geometrical size (length, radius), power consumption, and gas mixtures used. Applications of each jet are also briefly covered.



Introduction

One of the attractive features of non-thermal atmospheric pressure plasmas is the ability to achieve enhanced gas phase chemistry without the need for elevated gas temperatures. This attractive characteristic recently led to their extensive use in applications that require low temperatures, such as in material processing and in biomedical applications.[1–4] Various discharge sources capable of generating stable and relatively homogeneous plasmas at or around atmospheric pressure have been developed and characterized in recent years. Excellent reviews of these sources can be found in ref.[5–7]. Many of these plasma sources use a barrier-like approach and all of them produce plasmas that are either geometrically confined to the area between the electrodes or contained within some sort of chamber or containment enclosure. Although this is very useful in several applications, there are cases where it is more desirable if the plasma were launched outside to an area not bound by anything. Plasma jets or plumes fill exactly such a niche. In this paper, a variety of cold plasma jets will be reviewed. They are grouped and classified according to the type of power supply they use. The operational characteristics of each jet will be outlined and the applications for which they have been employed are mentioned. It is important at this point to mention to the reader that thermal jets, plasma torches, and laser-produced plasma plumes are not discussed in this review.

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Atmospheric Pressure Non-Equilibrium Plasmas

Many of the plasma jets/plumes reviewed in this paper are launched into the surrounding environment by devices that internally generate atmospheric pressure non-equilibrium plasmas (also called non-thermal plasmas, NTP). The jets/plumes are 'blown' outside the device by gases flowing at rates that are typically several liters per minute. For this reason, a brief description of atmospheric pressure non-equilibrium discharges (or NTPs) is now given. As a note to the reader, this is not a detailed review of NTP sources, as there is already an extensive literature comprising regular papers as well as topical reviews and general reviews of non-equilibrium atmospheric pressure plasmas, see ref.[5–12] and references therein. So, the following is just a very quick overview of the major generation methods since these same methods are employed as an integral part in the design of most of the cold plasma jets covered in this review paper.

NTPs have been generated under a wide range of driving frequencies and with various electrode geometries. One requirement is common to all the methods: the inhibition of the glow-to-arc transition. Various methods achieve this requirement by different means.

Using semiconductor-manufacturing techniques microdischarge structures were used to generate DC-driven plasmas.[13] Hollow cathode discharges with sub millimeter cathode dimensions have also been widely employed.[14] Microdischarges generate high-pressure non-thermal plasmas by having cathode dimensions in the micrometer range, which allows an increase in pressure while maintaining a relatively low sustaining voltage. DC and line frequencies (50/60 Hz) were also used to produce NTPs. In this case resistive barrier discharges which use a uniformly distributed resistive layer/film to cover the electrodes are employed.[15] Here the discharge current is limited by the resistive layer, which plays the role of ballast, and does not allow the discharge to transition to an arc.

One of the most prevalent devices to generate NTPs is the dielectric barrier discharge (DBD).[16] DBDs use dielectric materials to cover one or both of two electrodes, and use high voltages in the kHz frequency range to ignite the discharge. Both planar and cylindrical electrode geometries have been used.

In DBDs, charge collection on the dielectric layer that covers the electrodes causes a drop of the voltage across the plasma every time it is ignited and causes it to extinguish. Because of this DBDs are self-pulsed discharges that do not allow the discharge current to increase to a level that induces arcing. DBDs typically generate non-uniform plasmas that exhibit a large number of filaments/streamers that appear randomly between the electrodes. These filaments are nearly cylindrical plasma columns with sub-millimeter radii and a lifetime that is only a few nanoseconds long.[17] Under some special conditions (frequency range and gas mixture) DBDs can also generate uniform diffuse plasmas that are filament free.[18] This mode of operation of DBDs has in the past two decades been the subject of numerous investigations because of its usefulness in material processing that include biomedical applications.[19,20]

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Radio frequency (RF) sources have also been used to generate NTPs with devices that are similar to DBDs and with devices where the electrodes are bare metal.[21] With bare metal electrodes, arcing is a problem and one has to control both the temperature of the electrodes (typically by water cooling) and to adjust the gas flow rate to a certain level to minimize the risk of drawing arcs. RF driven devices require impedance matching between the power source and the plasma to optimize the power deposited in the plasma and minimize the reflected power. For detailed information on the modes of operation of RF sources the reader can refer to ref.[10–12].

The driving frequency has recently been extended to the microwave region (2.45 GHz).[22] In the microwave region the power is transported by a waveguide with a tuning section for impedance matching. The operating gas is injected in a nozzle located at a position (a quarter wavelength away from the shorted end of the waveguide) where the electric field strength is maximum.[22] In the device described by Park et al.[22] an inert gas such as argon at flow rates of several tens of liters per minute was used to generate an NTP discharge.

The plasma generation schemes described above have all been used as a basis on which cold plasma jets are designed. However, until now there has not been many attempts to investigate the physical processes that can elucidate the way jets/plumes are initiated and maintained in an atmospheric pressure environment that is, in most cases, simply the surrounding air. Most of the jets/plumes that will be described in this paper are ejected from a plasma source and into a region (room air) where the electric field can be very weak or even non-existent. So how low temperature plasmas can be maintained under such experimental conditions is a question worth delving into. Lu and Laroussi[23] proposed the following model to explain how the plume emitted by their plasma jet (the plasma pencil, a device driven by nanosecond high voltage pulses) is produced. This model may apply to many of the cold plasma jets/plumes used today.

The velocity at which the plasma plume travels was measured and found to be several order of magnitude greater than the gas flow velocity in the device, which is about 8 $\mathrm{m \cdot s^{-1}}$. To have a plume travel at high speed under very low electric field, photo-ionization has to play an important role. In the authors photo-ionization-based model[23,24] the head of a cathode-directed streamer is assumed to be a sphere of radius r_0, containing n^+ positive ions. As the streamer head moves forward, it leaves behind a quasi-neutral ionized channel with a very low conductivity; the head is not connected to the anode and only the streamer head is measurably luminous. This was also observed experimentally.[23] Therefore, the following sequence of events was proposed:[23]

Assume that at a given instant of time, the streamer head consists of a small sphere, which has a radius r_0 and a space charge n^+. Because of photon emission from the streamer, suppose that a single photoelectron is created at a suitable distance r_1 from the center of the sphere. Under the influence of the field set up by the space charge, the electron is accelerated towards the sphere and an avalanche is initiated. In moving towards the sphere, from r_1 up to some point r_2, the electron forms an avalanche of multiplication

$$n = \exp \int_{r_2}^{r_1} \alpha \mathrm{d}r, \tag{1}$$

and of diffusion radius

$$r_0 = \left(6 \int_{r_2}^{r_1} \frac{D}{v_\mathrm{d}} \mathrm{d}r \right)^{1/2}, \tag{2}$$

where α is Townsend's first ionization coefficient, D is the diffusion coefficient, and v_d is the electron drift velocity. If the multiplication up to the sphere is sufficient, the electrons neutralize the positive charge but leave behind a new positive region. The best value of r_1 could in principle be obtained from an exact knowledge of the number and type of photons emitted from the sphere within a particular solid angle, their absorption coefficients, and ionizing efficiency. Since complete data on these quantities is not available, r_1 is taken as the distance at which the electric field strength is such that ionization and attachment rates become equal. For air, this occurs when $E/p = 30\ \mathrm{V \cdot cm^{-1} \cdot mmHg^{-1}}$.[24]

According to Dawson and Winn,[24] the following three requirements must be fulfilled in order for the streamer propagation under low or zero field to occur: 1) The number of new positive ions created by the avalanche must be equal to n^+, i.e., the number of ions in the original sphere; 2) The diffusion radius of the avalanche head must not become larger than r_0; and 3) the avalanche must reach the required amplification before the two charge regions begin to overlap, i.e., $2r_0 \le r_2$.

Next, r_2 and r_0 are calculated for different values of positive charge n^+. The procedure is as follows: First, a value of n^+ is given. Second, the electric field as a function of r from simple electrostatics is calculated according to:

$$E = \frac{Q}{4\pi\varepsilon_0 r^2} \tag{3}$$

Under atmospheric pressure air conditions the main ion is N_2^+ even if the jet/plume contains a carrier gas (such as

Table 1. Calculated radii (r_o and r_2) for different original space charge numbers.[17]

Radius	n^+ (number of original positive charges) ($\times 10^9$)				
	1	2	3	4	5
r_2 (cm)	0.02	0.1	0.17	0.23	0.3
r_0 (cm)	0.056	0.068	0.075	0.080	0.085

helium).[25] In addition, the attachment process is dominated by O_2, also from air, which diffuses to the plume. The distance r_1 is determined as the distance at which the reduced electric field is equal to 30 $V \cdot cm^{-1} \cdot mmHg^{-1}$.[24] r_2 is then calculated according to Equation (1) when n is equal to n^+. The Townsend's first ionization coefficient α is calculated according to:[26]

$$\alpha = 15p \cdot \exp(-365 p/E)_{\mathrm{cm}^{-1}} \tag{4}$$

where p is the pressure and air is assumed to be the background gas. Finally, by using Equation (2), r_0 is calculated, where D and μ_e were determined according to:[26]

$$D = \frac{2 \times 10^5}{p[\mathrm{torr}]} \mathrm{cm}^2 \cdot \mathrm{s}^{-1} \tag{5}$$

$$\mu_e = \frac{0.86 \times 10^6}{p[\mathrm{torr}]} \mathrm{cm}^2 \cdot \mathrm{V}^{-1} \cdot \mathrm{s}^{-1} \tag{6}$$

Table 1 shows the calculated r_2 and r_0 for different n^+.[23] In these calculations, the values of D and v_d were determined for the case of helium gas. The three requirements for the streamer self-propagation are met only if $2r_0$ is smaller than r_2. From Table 1, it can be seen that when n^+ is less than 2×10^9, $2r_0$ is greater than r_2. This means that the streamer head can't self propagate under this condition. However, when n^+ is larger than 3×10^9, $2r_0$ is smaller than r_2, meaning that the streamer head can self propagate under low or zero external electric field. Therefore, according to Dawson and Winn,[24] the plume velocity can reach values as high as 10^6 $m \cdot s^{-1}$ and it can travel up to several centimeters without the presence of an external electric field. This is in agreement with the experimental observations.[23]

Atmospheric Pressure Cold Plasma Jets

The plasma jets described in this section are classified according to an ascending frequency range of the power supplies used to ignite and sustain them. These frequencies range from DC (or pulsed DC) to the GHz.

A. Pulsed DC-Driven Plasma Jets

Several plasma jet approaches that use pulsed DC power have been developed. Voltages in the kV range, pulse widths that range from the nanosecond to the microsecond at kHz repetition rates, and argon, helium, nitrogen, or a mixture of these with oxygen have been used.

Duan et al.[27] developed a plasma jet comprised of a hollow Teflon tube equipped with two planar electrodes (inside the tube) with the operating gas or gas mixture flown inside the tube. To avoid arcing, the electrodes are ballasted (for current limitation). With a gas flow the plasma can be blown outside the tube to form what the authors called a 'plasma brush'. The authors claim that they used both continuous DC and pulsed DC successfully. The plasma power consumption is limited to a few watts and the gas temperature was measured to be in the 40–85 °C range for gas flow rates that range from 3 500 to 1 000 sccm, respectively.

Forster et al.[28] developed a plasma jet by using a DBD configuration. In their device the ground electrode was a brass rod placed in the middle (along the axis) of a dielectric tube made out of fused silica. The high voltage electrode was a brass ring wrapped around the dielectric tube. The gap distance between the rod electrode and the dielectric tube inner surface was 0.7 mm. Figure 1 is a schematic of the device. By flowing argon at a flow rate of 250 $L \cdot h^{-1}$ and applying high voltage pulses (15 kV high and 600 ns wide at a repetition rate of 25 kHz) they were able to generate a plasma jet. The gas reached temperatures up to 100 °C but the tip of the jet could be touched by bare hands (see Figure 2). The authors claim that using this plasma jet they were able to increase the surface wettability of wood.

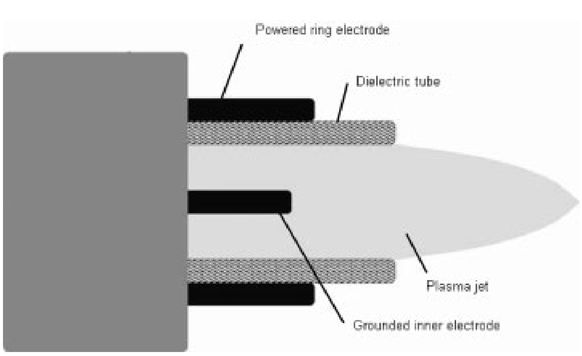

Figure 1. Schematic of the plasma jet developed by Forster et al.[28]

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Figure 2. Photograph of the Forster et al.'s jet.[28]

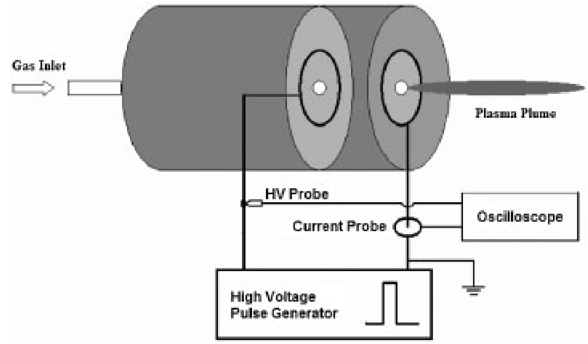

Figure 3. Schematic of the plasma jet developed by Laroussi and Lu.[25] The electrodes are circular copper rings attached to centrally perforated alumina disks. The plume, which can reach lengths in excess of 50 mm, is launched from the central orifice (3 mm in diameter). The voltage pulses are 500 ns wide with repetition rates of few kHz.

Zhang et al.[29] also reported on a plasma jet similar to the one described above except that the dielectric tube was a quartz capillary and the inner electrode is a tungsten rod/wire. In this case the inner electrode was connected to the high voltage source and the outer loop electrode was grounded. The applied voltage pulses were in the 1–15 kV range in magnitude with a repetition rate of 15 kHz. Argon, helium, and nitrogen were all used as operating gas, however, the longest jet/plume (44 mm long) was achieved when helium was used at a gas velocity of 20 $m \cdot s^{-1}$. The authors found that there is a critical gas velocity beyond which a jet can be formed. For helium, argon, and nitrogen these critical velocities are 3, 5, and 8 $m \cdot s^{-1}$, respectively.

Walsh et al.[30] carried out a comparison between a DBD-based plasma jet powered by a sinusoidal voltage source and by a pulsed power source. The device consisted of a dielectric tube with a metal belt/ring electrode wrapped around it. The ground electrode was a metal plate placed at a distance (3–5 cm) from one end of the dielectric tube. When helium was flown through the tube at a flow rate of 5 $L \cdot min^{-1}$ a jet reaching out to the grounded plate was produced. In the sinusoidal excitation case the applied voltage was 7.3 kV peak-to-peak and the frequency was in the 1–10 kHz range (nominally at 7 kHz). In the pulsed excitation case a unipolar pulse train with pulses 71 μs wide and 4 kV high were used. The conclusion of the comparison was that the pulsed case was much more efficient energetically as it took 12 times more energy in the sinusoidal case to generate the same concentration of atomic oxygen, atomic oxygen being an important species that plays a crucial role in surface oxidation processes.

Laroussi and Lu[25] developed a hand-held plasma jet device that they called a 'plasma pencil'. This device is conceptually very different from the jet devices described above. It consists of two electrodes, each made of a thin copper ring attached to the surface of a centrally perforated alumina (Al_2O_3) disk. The hole in the center of the alumina disk is 3 mm in diameter, while the diameter of the disk is about 2.5 cm. The diameter of the copper ring is greater than that of the hole but smaller than that of the disk. The two electrodes are inserted in a dielectric cylindrical tube of about the same diameter as the disks and are separated by a gap the distance of which can be varied in the 0.3–1 cm range. Figure 3 is a schematic of the device. The two electrodes are connected to a high voltage pulse generator capable of producing pulses with amplitudes up to 10 kV, pulse widths variable from 200 ns to DC, and with a repetition rate up to 10 kHz. The rise and fall times of the voltage pulses are about 60 ns.

When a gas such as helium or argon is injected at the opposite end of the dielectric tube and the sub-microsecond high voltage pulses are applied to the disk electrodes, a discharge is ignited in the gap between the electrodes and a plasma plume/jet is launched through the hole of the outer electrode and into the surrounding room air. Figure 4 is a photograph of the device in operation. Laroussi and Lu reported jet/plume lengths of up to 5 cm and a plume gas temperature of 290 K.[25] The

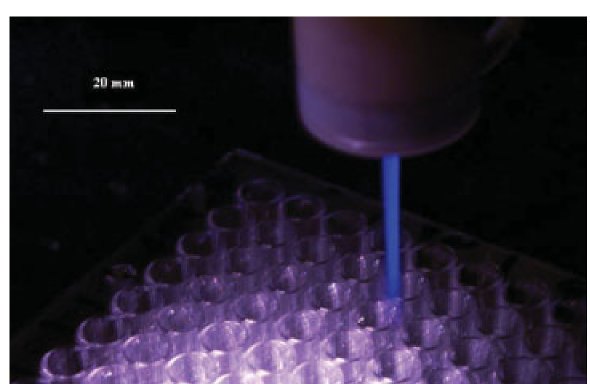

Figure 4. Photograph of the plasma pencil in operation.[31]

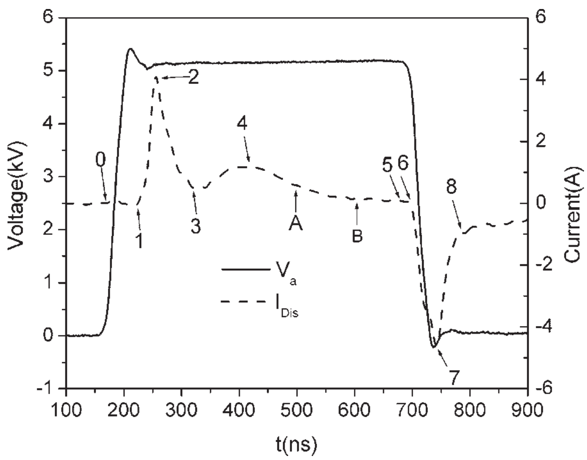

Figure 5. Current–voltage characteristics of the plasma pencil.[23]

current–voltage (*I–V*) characteristics of the device show that three current pulses occur per single voltage pulse. The authors claim that the second pulse, which appears about 100 ns after the first, is directly related to the launching of the plasma plume out of the exit aperture of the device. Figure 5 shows typical *I–V* characteristics. In a subsequent publication,[23] these authors reported that the plasma plume was actually a train of small plasma bullets traveling at very high speeds (up to 100 $km \cdot s^{-1}$ as

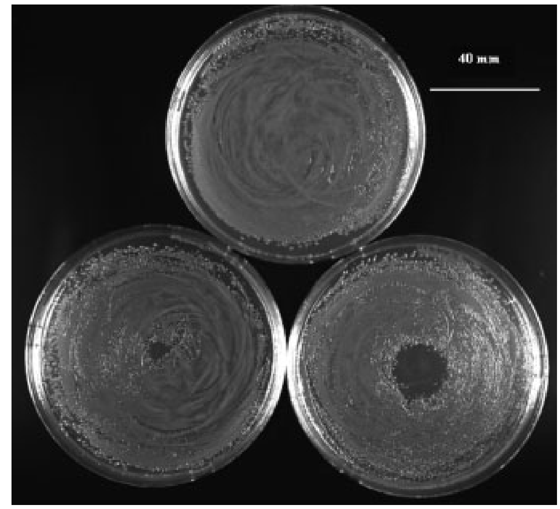

Figure 7. Localized inactivation of *E. coli* by the plasma pencil.[31] The top petri dish is the control, the left and right petri dishes represent 30 and 120 s plasma exposures, respectively. Helium is the operating gas.

compared to the gas flow velocity of only 8 $m \cdot s^{-1}$). Figure 6 is a series of high-speed photographs taken by an ICCD camera showing the plasma bullet at different spatial locations. Laroussi et al.[31] used the plasma pencil for biomedical applications to demonstrate its potential for killing various types of bacteria. Since the cross-section of the plasma plume is small, the treatment of samples is localized. This permits a targeted means to selectively treat surfaces. Figure 7 shows an example of how a localized zone treated by the plasma pencil remains free of colony forming units of *Escherichia coli* bacteria after a period of incubation.

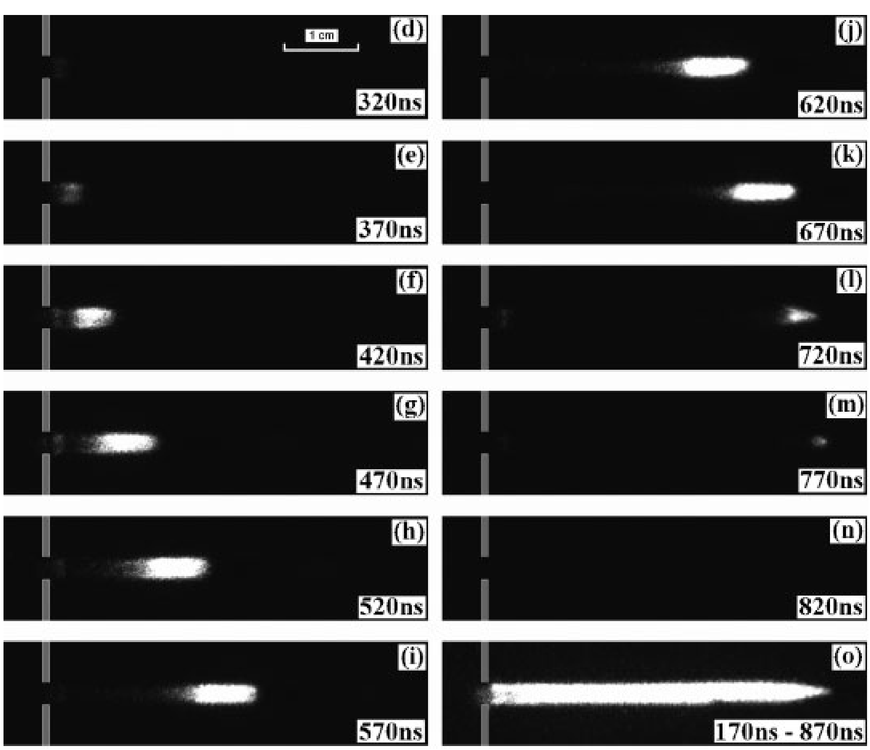

Figure 6. ICCD photographs of the plasma bullet emitted by the plasma pencil.[23] The plasma bullet is launched about 250 ns after the rising edge of the voltage pulse. The exposure time of each photograph is 50 ns.

B. AC-Driven Plasma Jets

Teschke et al.[32] reported on a plasma jet generated by a DBD-like device. The device consists of a dielectric tube with two tubular electrodes mounted on it. A sinusoidal high voltage of a few kV at frequencies in the 5 to 50 kHz range is applied between the electrodes and a gas (helium) is flown through the tube at a velocity of 16.5 $m \cdot s^{-1}$. Using an ICCD camera the authors reported that the plume consists of plasma bullets traveling at a velocity of 15 $km \cdot s^{-1}$. This bullet velocity is

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lower than that reported by Lu and Laroussi.[23] It appears that the fast rising voltage pulses (60 ns rise time) used by the latter authors are responsible for the higher bullet speeds. Teschke et al.[32] also reported that the structure of the bullet was not homogeneous in volume. They observed that the center was very luminous as compared to the outer edges. They claim that the reddish intense point in the center of the bullet is a result of emission from helium while the outer blue/white light comes from the nitrogen, which is supplied by the surrounding air.

Cheng et al.[33] developed a cold plasma jet to treat the surface of poly(propylene) (PP) and poly(ethylene terephthalate) (PET). The device is made of two concentric metal tubes with the outer tube connected to the high AC voltage and covered by a dielectric layer. Figure 8 is a schematic of the device. Voltages of 30 to 80 kV are applied between the electrodes at frequencies in the 6 to 20 kHz range. Argon is blown through the inner tube electrode (grounded) at flow rates in the 50 to 2 500 $L \cdot h^{-1}$ range. The jet forms at the nozzle of the device with temperatures between 25 and 30 °C. Figure 9 is a photograph of the plasma jet. The authors reported that the wettability of PP and PET had substantially increased after only 5 to 10 s exposure to the plasma jet. For PET the water contact angle decreased from 150° to 0°. This is because the water completely infiltrated the PET fibers. For PP the water contact angle decreased from 120° to 60°. This was a limit that could not be surpassed even for longer treatment times.

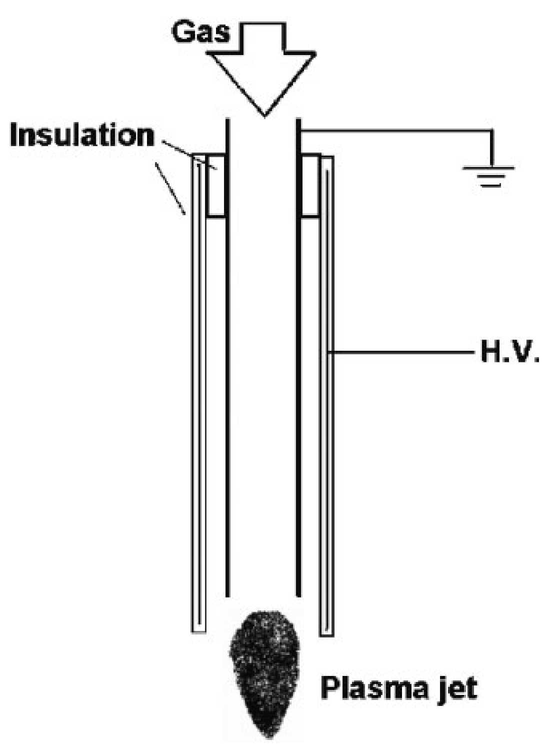

Figure 8. Schematic of the plasma jet developed by Cheng et al.[33] Voltages of 30 to 80 kV at frequencies that range from 6 to 20 kHz are applied between the concentric electrodes.

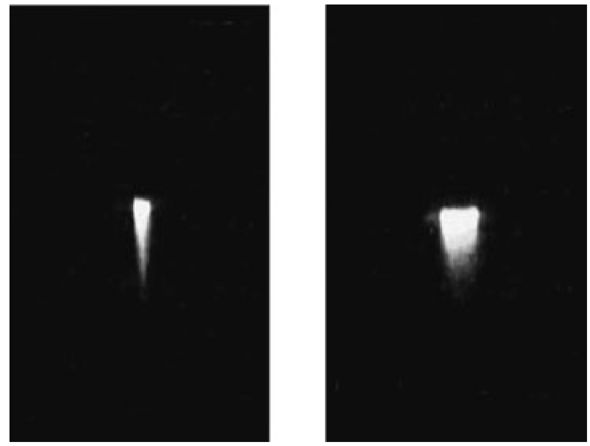

Figure 9. Photographs of Cheng et al.'s jet under different operating conditions.[33] The picture to the left is for a 4 mm diameter nozzle while that on the right is for 20 mm diameter nozzle.

Chen et al.[34] developed what they called an 'APGD plume' for the degradation of organic contaminants. It is a DBD-based design where a quartz condenser tube was used as the dielectric layer. A potassium chloride (KCl) solution in the outer layer of the reactor acted as the outer electrode. It was connected to a water pump and a solution tank 25 cm away from the plasma reactor outlet. The conductive liquid in the outer layer, which contacted the dielectric layer, also served as coolant for the system. The second electrode (connected to the high voltage) was a metal rod centered inside the condenser tube by Teflon and ceramic fittings. The gap distance between the rod electrode and the inner surface of the quartz tube was 2 mm. The applied voltage was up to 30 kV at frequencies that ranged from 8 to 30 kHz. The tapered end of the tube acted as a nozzle and allowed for the launching of a plasma plume, the temperature of which did not exceed 320 K in the power range of 5 to 50 W. Argon or helium were used as the operating gas at a flow rate of 0.8 $m^3 \cdot h^{-1}$. The plume generated by the device is 10 mm in diameter with lengths in the 7 to 40 mm range. Figure 10 is a photograph of the plasma plume. This plasma jet was used for the degradation of methyl violet 5BN (MV-5BN). It was found that it was the excited species that came from the nitrogen and oxygen in the surrounding air that caused degradation of the sample. The authors claim that the UV generated by the plasma plume did not play a significant role in the degradation process. In addition the authors claim that only the downstream oxygen was able to break the aromatic ring structure of the dye molecules.

Kim et al.[35] developed what they called a 'corona DBD hybrid type discharge' that can generate a microjet. The electrode system is basically a needle to dielectric plane

Figure 10. Photograph of the APGD plume developed by Chen et al.[34] The outer electrode is a solution of potassium chloride. The second electrode is a centrally located metal rod. The applied voltage is 30 kV at frequencies that range from 3 to 30 kHz.

electrodes. Using voltages in the 0–2 kV range with a frequency between 30 and 70 kHz and a helium flow rate between 5 and 25 standard liters per minutes (slpm) they were able to produce a jet the characteristics of which can be adjusted simply by adjusting two geometrical parameters, namely the distance between the tip of the needle and the end of a quartz sleeve (where the needle is placed), *s*, and the distance between the tip of the needle and the surface of the plane electrode, *d*. These distances (*s* and *d*) are typically 2 and 5 mm, respectively. The gas temperature was reported to be in the 300–490 K range depending on the type of plasma generated, which is controlled by the parameters *s* and *d*. The authors claim that in a preliminary test this jet was able to change the wettability of a thin polymer film. (The authors did not specify the kind of change and the type of polymer used.)

C. RF-Driven Plasma Jets

There is a relatively large number of RF-driven jets that have been developed through the years. We are not going to cover each one of them as this will lead to redundancy. Instead only a few jets will be discussed. The choice of these devices is motivated by their originality and/or by the large amount of work that was dedicated to them.

Koinuma et al. developed the earliest RF cold plasma jet, which they reported in *Applied Physics Letters* in 1992.[36] Since then this group published several papers describing variant designs of their device and its applications.[37–43] The device, which the authors named a ‘microbeam plasma generator’, used as a cathode a needle electrode (tungsten or stainless steel with 1 mm diameter) connected to an RF source (13.56 MHz). The needle electrode was placed inside a quartz tube, which was surrounded by a cylindrical anode connected to ground. Figure 11 is a schematic of the microbeam plasma generator. Using helium with a typical flow rate of 70 sccm, a plasma ‘beam’ (2 mm in diameter and up to 5 mm long) could be generated and launched into the open air. In later work the authors used other materials for the needle and the dielectric tube. These were platinum for the needle electrode and alumina for the dielectric tube. Koinuma and co-workers used this device for various material processing applications. Depending on the application, helium or argon were mixed with various gases. This led to different characteristics of the beam/jet produced and especially to different plasma temperatures. So temperatures ranged from quite low (with helium) to up to 450 °C for argon mixed with reactive gases in film deposition applications. As mentioned earlier, the microbeam plasma generator was used by Koinuma and co-workers in many material processing applications.[37–43] Amongst these is the surface treatment of a vulcanized rubber compound to improve its adhesion properties,[42] and the etching of silicon, Si (100),

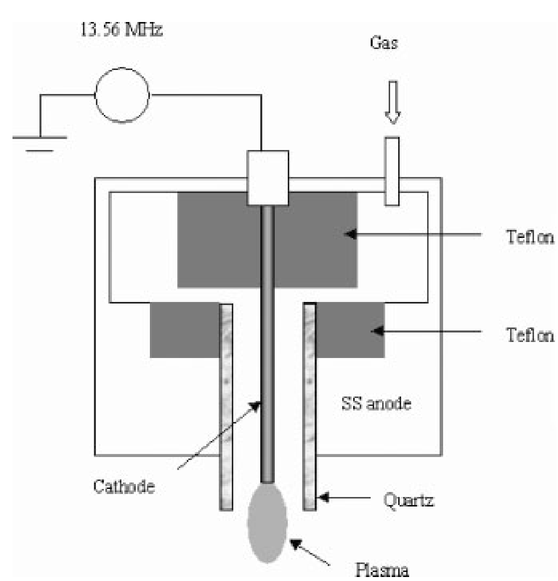

Figure 11. Schematic of the microbeam plasma generator developed by Koinuma et al.[36] This is one of the first reported cold plasma jets. The cathode is a needle to which RF power at 13.56 MHz is applied.

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by using a stainless steel needle cathode and an operating gas mixture of 1% CF_4 in a balance of helium.[36] Using argon with small amounts of oxygen, photoresist ashing at a rate greater than 1.2 $\mu m \cdot min^{-1}$ was achieved.[37] The temperature of the plasma in this application was 240 °C. Deposition of silicon dioxide, SiO_2, films on various substrates (Si (100) wafer, Si_3N_4 plate, Cu plate, and corning glass 7059) at deposition rates greater than 10 $nm \cdot s^{-1}$ using tetramethoxysilane, TMOS (0.2–l sccm) carried with Ar (5– 25 sccm) and H_2 (0–5 sccm) into argon plasma was achieved. Other applications such as the deposition of TiO_2 and the production of fullerenes were also conducted.[40,43] The authors also predicted that their microbeam plasma may be useful in biological applications.[36]

The atmospheric pressure plasma jet (APPJ)[44] is a capacitively coupled device that consists of two co-axial electrodes between which a gas flows at high rates. Figure 12 is a schematic of the APPJ. The outer electrode is grounded while the central electrode is excited by RF power at 13.56 MHz. The free electrons are accelerated by the RF field and enter into collisions with the molecules of the background gas. These inelastic collisions produce various reactive species (excited atoms and molecules, free radicals, etc) which exit the nozzle at high velocity. Figure 13 is a photograph of the APPJ in operation. The developers of the APPJ carried out extensive diagnostics and/or calculations to measure/estimate important parameters such as temperature, and concentrations of active species and charged particles. They report that the electron density is in the 10^{11} cm^{-3} range, a jet temperature that varies from 100 to 275 °C depending on the amount of RF power dissipated by the device, an ozone concentration in the 10^{15} cm^{-3} range, an atomic oxygen concentration of 8×10^{15} cm^{-3} at the nozzle exit, an oxygen metastable (singlet Δ) concentration of 2×10^{13} cm^{-3} at the nozzle exit, and an ion flux in the 10^{13} cm^{-2} range.[45–50]

The APPJ was used for various processing applications, including biological.[39–44] For example, inactivation of spores of *Bacillus globigii* (BG spores) with a decimal value

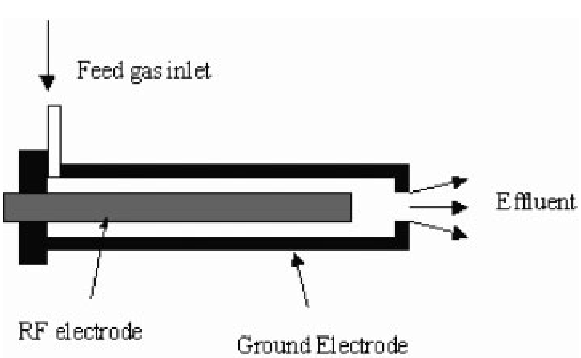

Figure 12. Schematic of the atmospheric pressure plasma jet (APPJ).[42] The APPJ is an RF-driven plasma jet.

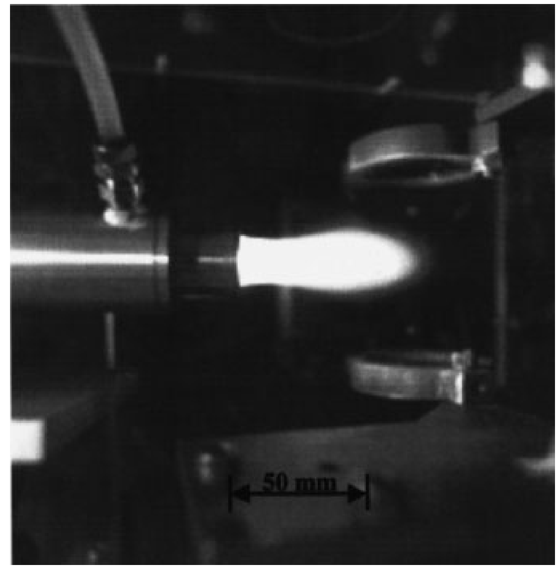

Figure 13. Photograph of the APPJ in operation.[48]

(time to kill 90% of the initial spore population) of 4.5 s using air as the operating gas was achieved.[45] The APPJ was also able to destroy sulfur mustard gas (a simulant to VX gas, which can be used in chemical warfare).[45] Using an admixture of 2% CF_4 to a mixture of oxygen and helium, the APPJ was used successfully to etch SiO_2, tantalum (Ta), kapton, and tungsten (W).[49] The etching rates achieved were 8 $\mu m \cdot min^{-1}$ for kapton, 1.5 $\mu m \cdot min^{-1}$ for SiO_2, 2 $\mu m \cdot min^{-1}$ for tantalum, and 1 $\mu m \cdot min^{-1}$ for tungsten. Mixing He and O_2 with tetraethoxysilane, SiO_2 films were deposited on Si (100) substrates using the APPJ.[50]

Janca et al.[51] developed an RF-driven (13.56 MHz) plasma jet that they named the HF plasma pencil. Its core is a pencil-shaped dielectric tube with a built-in hollow electrode. This electrode is basically a thin pipe with an inner diameter of 1–2 mm. The RF power applied to the hollow electrode is adjusted in order to obtain a torch-like plasma at the edge of the electrode. The working medium can be a gas, a liquid, or a powder. The authors report that the neutral gas temperature reaches up to 7×10^3 K. They also report an electron number density of 2×10^{13} cm^{-3}. The HF plasma pencil/torch has been used in several applications that range from the production of fullerene, polymerization in liquids, the cleaning of archeological glass artifacts, etc.[51]

Stoffels et al.[52–62] developed an RF plasma jet that they called the 'plasma needle' and which they used extensively in biomedical applications. In the early version of the device the powered electrode was a needle-like stainless steel wire 5 cm long and 1 mm in diameter located in the center of a metal grounded cylinder. The cylinder was filled with helium but was not airtight. In a later version of the device the needle electrode was insulated by a glass sleeve but with a protruding tip. The insulated needle was put

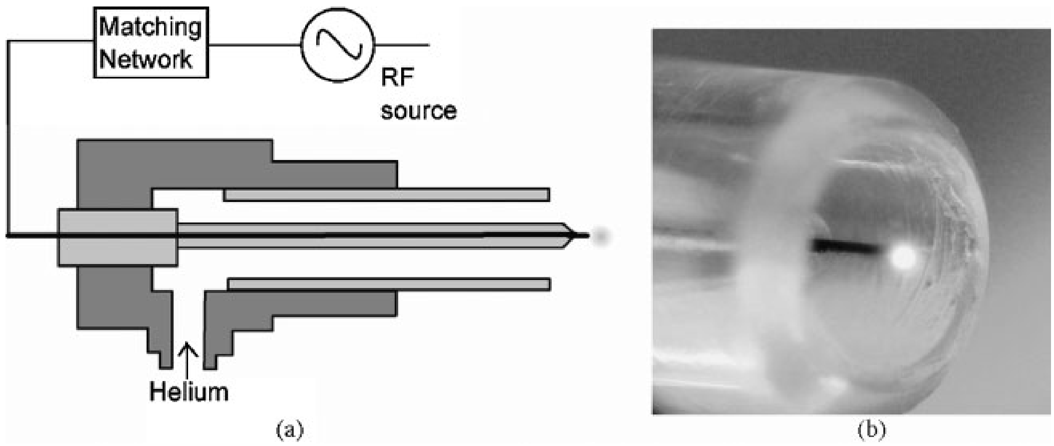

Figure 14. Schematic (a) and photograph (b) of the plasma needle developed by Stoffels et al.[58] The plasma needle is RF driven and has been used extensively in biomedical applications.

coaxially in a Perspex tube where helium was flowing at the rate of 2 $L \cdot min^{-1}$. The entire structure was fixed in a stainless steel holder. The needle electrode consisted of a metal alloy pin (0.3 mm diameter). The metal pin protruded from the stainless steel holder by 1.5 cm, while the total length of the needle was about 8 cm. Using RF power a small plasma plume (0.1 mm in diameter and up to 2 mm in length) could be generated at the tip of the needle electrode. In various experiments the authors used RF frequencies of 13.05, 13.56, and 7.17 MHz and RF powers that ranged from 10 to 300 mW. Figure 14 is a schematic and a photograph to illustrate the device and the visual appearance of the discharge.[58]

Various diagnostics were used to characterize the plasma. Using mass spectrometry it was found that the plasma needle is a good source of NO.[59] Optical emission spectroscopy showed the presence of atomic oxygen, hydroxyl radicals, and N_2^+ ions in the plasma. Ultraviolet radiation was also emitted with the strongest emission in the 305–390 nm wavelength range. With helium as the operating gas, the plasma temperature was measured to be close to room temperature and the electrons temperature was estimated to be in the 0.2–0.3 eV range.

Stoffels and co-workers used the plasma needle in various biomedical experiments. They showed that the plasma generated by their device can be used to kill bacteria, to manipulate eukaryotic cells (such as in cell detachment without causing necrosis), and to induce apoptosis in 3T3 mouse fibroblast cells.[57–62] In addition, treatment of mammalian vascular cells that are prevalent in arterial walls cells was tested. Two cell types were used in the experiments: endothelial cells and smooth muscle cells. The specific cell types were Bovine aortic endothelial cells (BAEC) and rat aortic smooth muscle cells. It was found that exposure as short as 10 s induced cell detachment generally without necrosis.

Foest et al.[63] developed two versions of an RF (13.56, 27.12, or 40.78 MHz) plasma jet. The first used two ring electrodes (one powered and the second grounded) wrapped around a quartz capillary tube (1 mm in inner diameter) where the operating gas flows. The second used a single ring electrode around a glass capillary (2.5 mm in inner diameter) and a rod electrode centrally introduced inside the capillary tube. The rod electrode was RF powered while the ring electrode was grounded. Figure 15 is a schematic of both devices. The second device can be better operated using molecular gases while the first was used with rare gases only. To provide for an independent introduction of precursor gases (for thin films deposition applications) additional channels can be added. This lowers the likelihood of precursor degradation and prevents the coating of the tip of the rod electrode.

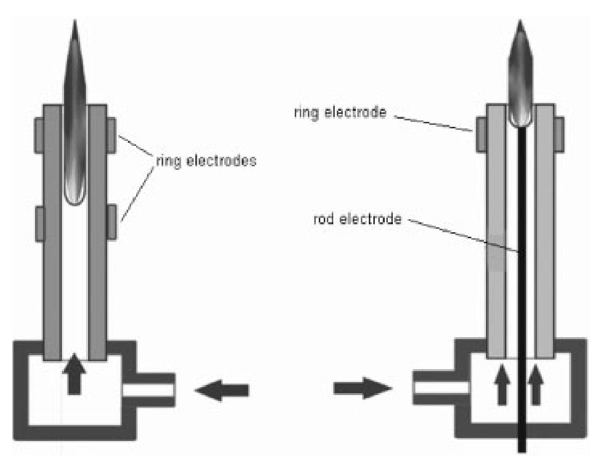

Figure 15. Schematics of both versions of the RF plasma jet developed by Foest et al.[63] The device to the left uses two ring electrodes while the device to the right used one ring electrode and a centrally introduced rod electrode. This jet is RF driven.

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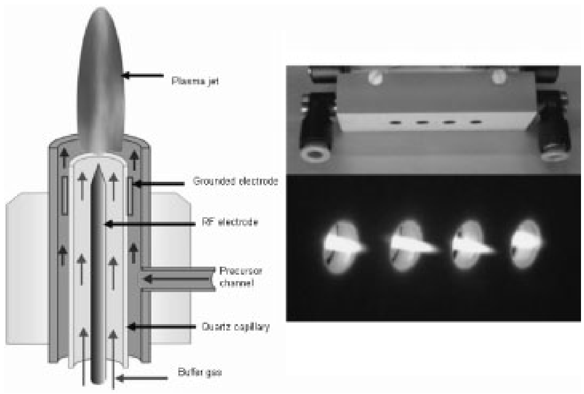

Figure 16. Schematic of Foest et al.'s device with a precursor channel and photograph of multiple jets based on that design.[63]

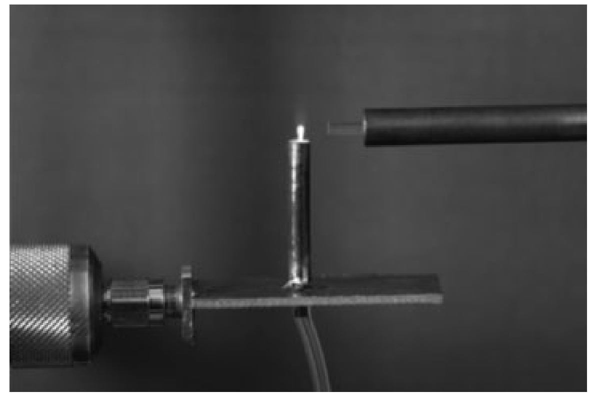

Figure 17. Photograph of the microwave plasma jet developed by Stonies et al.[73] The plasma is ignited on top of an open-ended coaxial line into which the operating gas is introduced. Jets of up to 4 mm in length are generated.

Figure 16 shows a schematic of a device with a precursor channel and a photograph of an apparatus based on that design. This device was used in a thin film deposition experiment using argon or nitrogen as a carrier gas (flow rates in the 3–10 slpm range) with admixtures of a few ppm of silane (SiH_4) in one experiment and two silicon–organic precursors hexamethyldisiloxane ($(CH_3)_3$–Si–O–Si–$(CH_3)_3$, HMDSO) and ($(C_2H_5O)_4$–Si, TEOS) in the permil range in another experiment. Applications of SiO_x-like films deposited by this device are the increase of scratch-resistance of polymeric materials and enhancement of barrier properties against gases, polyolefins, or water.[63]

In addition to the RF-driven devices described above, many others have been developed. The reader can find more information about these in ref.[64–72].

D. Microwave-Driven Plasma Jets

Based on a scaled-down version of a microwave plasma torch (MPT), Stonies et al.[73] developed a small atmospheric pressure plasma source using microwave power at 2.45 GHz and low power levels (down to 2 W). The plasma was ignited on top of an open-ended coaxial line with an inner capillary where the operating gas is passed. A microstrip line was used to match the plasma impedance to that of the microwave source. Figure 17 shows a photograph of the system. Argon gas with flow rates down to 70 mL $\cdot$ min^{-1} was used and jets up to 4 mm in length were generated. Helium was also used successfully. This microwave driven source has applications in gaseous species detection and as an element specific detector in gas chromatography.[73]

Conclusion

Cold plasma jets/plumes emitted in atmospheric pressure environments constitute a very practical technology that can be used in various industrial and scientific applications. In this paper an extensive review of the various devices that have been developed in the last few years is presented. The devices are grouped according to the frequency of their power supply. Besides the description of the various generation methods, the potential applications of each device are also mentioned. It is the hope of the authors that researchers and engineers working in this field find the necessary initial information on the various cold plasma jets in the one single document that this paper represents.

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