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Viewpoint

The development of uniform atmospheric pressure glow discharges: appearance of stable glow discharge in air, argon, oxygen and nitrogen at atmospheric pressure using a 50 Hz source (S Okazaki *et al* 1993 *J. Phys. D: Appl. Phys.* 26 889)

Mark J Kushner
Electrical Engineering and Computer Science, University of Michigan, 1301 Beal Ave., Ann Arbor, MI 48109-2122, USA
E-mail: mjkush@umich.edu

This Viewpoint relates to an article by S Okazaki *et al* (1993 *J. Phys. D: Appl. Phys.* **26** 889) and was published as part of a series of Viewpoints celebrating 50 of the most influential papers published in the Journal of Physics series, which is celebrating its 50th anniversary.

The development of atmospheric pressure plasmas (APPs) for chemical synthesis, materials processing, environmental cleanup and now for biotechnology has been continually challenged by the desire, and the need, to produce uniform glow discharges. A scaling law in low temperature plasmas states that if ionization occurs only from the ground state, the gas density N is constant, and the E/N (electric field/gas density) is constant, the product $N\tau$ is constant, where τ is the characteristic time that the plasma will come into a steady state—or become unstable [1]. APPs, by virtue of their high pressure (large N), have small values of τ. This instability formation time is typically so short at atmospheric pressure that feedback control systems cannot respond quickly enough to prevent an instability. The end result is that strategies for obtaining stable and uniform plasmas in APPs have typically relied on passive techniques where active intervention by the user is not required.

The most common type of instability in APPs is the formation of an arc in which the discharge collapses into a small hot, intense region of plasma which often damages the electrodes. One of the features of APPs is that they are non-equilibrium, which means that the electron temperature can be controllably higher than the ion or gas temperature. This non-equilibrium nature of the plasma enables selective production excited states, ions and photons. The high temperature of the arc works against this selectivity by producing an equilibrium set of conditions, not unlike combustion. Although arcs are highly desirable in many applications, for low gas temperature, selective processing, non-thermal plasmas are usually called for. The first step towards producing uniform APPs was the prevention of arcs.

Perhaps the first example of a passive technique to stabilize an APP and prevent arcs is the dielectric-barrier-discharge (DBD) [2], invented by Ernst Werner von Siemens in 1857 as a means to produce ozone. The DBD is passively stable by inserting capacitance in series with the current flowing through the plasma—the dielectric barrier. By charging this series capacitance, the current through the plasma is cut off prior to an arc forming. Although the DBD is stable, it is usually not a uniform glow discharge. The plasma in a DBD is usually composed of a forest of small filaments having diameters of hundreds of microns [3]. Although this filamentary discharge serves well in many applications, it is often also desirable to have a uniform glow discharge. This means that on a microscopic basis, the plasma is spatially uniform without filaments, gradients or other inhomogeneities.

In their article, *Appearance of stable glow discharge in air, argon, oxygen and nitrogen at atmospheric pressure using a 50 Hz source*, Satiko Okazaki, Mashuhiro Kogama, Makoto Uehara and Yoshihisa Kimura addressed this technological challenge—the desire and need

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for uniform glow discharges at atmospheric pressures [4]. In doing so, they established operating principles for obtaining uniform glow discharges that pre-staged developments for the following nearly 25 years.

At the time, it was common practice to operate DBD-like electric discharges with low frequency voltages—50 Hz in the case of Okazaki *et al*. Unfortunately, the low frequency worked against producing homogeneous discharges from two perspectives. First, low frequency equates to low values of dV/dt (time rate of change of voltage). Our current understanding of DBDs teaches us that optimum glow-like performance can be obtained by operating with large values of dV/dt to enable the applied voltage to, at least momentarily, exceed the quasi-equilibrium operating (or breakdown) voltage, V_0 [5]. This scaling for dV/dt originates from there being a finite time that the discharge will transition into a conducting plasma, a time called the formative time-lag. If dV/dt is high enough, then a voltage higher than V_0 can be applied in a time shorter than the formative lag time. The end result is more rapid ionization and a more uniform discharge. Second, a low frequency means that there is a long period between discharges which allows the electron and metastable densities to decay to small values. Again, our current understanding of DBDs teaches that glow-like performance can be obtained by operating with reasonably large electron and/or metastable densities surviving from the previous pulse at the time of the next pulse [6].

Okazaki *et al* were limited to using small values of dV/dt by the availability of their hardware, and so devised a passive means to produce a distributed stabilizing effect. This was achieved by using combinations of a fine wire mesh and dielectric sheets as one of the electrodes. The distributed impedance of the fine wire mesh was successful at producing atmospheric pressure glow-like discharges by preventing hot-spots from occurring. This breakthrough enabled a wholly new regime of APP operation. In subsequent work, Kogoma and Okazaki demonstrated improved efficiency for ozone production when reducing the gap size with mesh electrodes, an effect typically not seen with conventional metal plate electrodes [7].

The work of Okazaki *et al* motivated a wide range of research into methods to produce uniform glow discharges, from high-repetition-rate discharges sometimes using seeded gas mixtures [8, 9] to the aforementioned use of short ns pulses [5]. Perhaps the most direct extension of the distributed impedance used in the devices of Okazaki *et al* is the work of Laroussi *et al* [10]. They replaced the wire mesh-dielectric electrode with a high resistivity but finite conductivity electrode. This configuration performs *distributed self-ballasting*. This is a concept that was developed in the early days of pulsed discharge lasers. The electrode in the early discharges used for CO_2 lasers consisted of a line of pins each of which was connected to the power supply through a resistor [11]. Should one of the pins collect a larger than average current, there is a larger voltage drop across the series resistor, which then locally reduces the voltage across the discharge thereby reducing the current. Laroussi *et al* extended the concept to large area, uniform glow discharges at atmospheric pressure, powered by both dc and radio frequency voltages. These discharges were then applied to sterilization of surfaces [12]. A similar concept using graphite electrodes was also used to stabilize early pulsed CO_2 laser discharges [13].

In conclusion, a recurring theme in the development of atmospheric pressure discharges is producing uniform, quasi-dc plasmas that provide controllable production of reactive species or uniformly treat surfaces. One of the early successes in achieving this goal was the innovative wire mesh electrode structures developed by Okazaki *et al* [4] for dielectric barrier discharges. Their innovations seeded many subsequent investigations that have produced the reliable, atmospheric pressure discharges in use today for applications from biotechnology to surface treatment.

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