Development of an Atmospheric-Pressure Plasma Jet for Thin-Film Sintering

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Abstract:

Atmospheric-pressure plasma jets (APPJ's), which generate plasma in open space rather than in a confined discharge gap or vacuum chamber, have recently become a topic of great interest. APPJ's have various applications such as surface modification, nanoparticle fabrication, and thin-film sintering. This sintering process can be applied to printed materials to increase their conductivity. For industrial applications, the major advantages for APPJ use are the system flexibility, ease of installation, and time and energy savings due to the fact there is no need for large furnaces or vacuum systems. This exploratory work focuses on developing an experimental APPJ setup using a dielectric barrier discharge (DBD) that can be used to sinter thin-film samples. Previous literature is reviewed, and various plasma jet geometries are compared. A final design is established, and sintering with graphene and silver samples is attempted. Some qualitative results are examined. Lastly, an evaluation of this technology and future design modifications are discussed.

Design Review:

Comparing Geometries

Over the past years, various successful APPJ geometries have been developed and refined, and there is a plethora of scientific research on the subject. With this project having such a large design space, it was important to first examine this documentation to get an idea of where to start. While some geometries are designed for very specific uses, there are a few general models that were considered for this project. Three primary geometries considered were: ring to ring, inner electrode to base, and inner electrode to ring. Also, the following are assumed to be using AC power supplies. As described by Bartis [1], a ring to ring configuration features two cylindrical electrodes that encompass a quartz tube, which houses flowing gas. An example setup is shown in Figure 1.

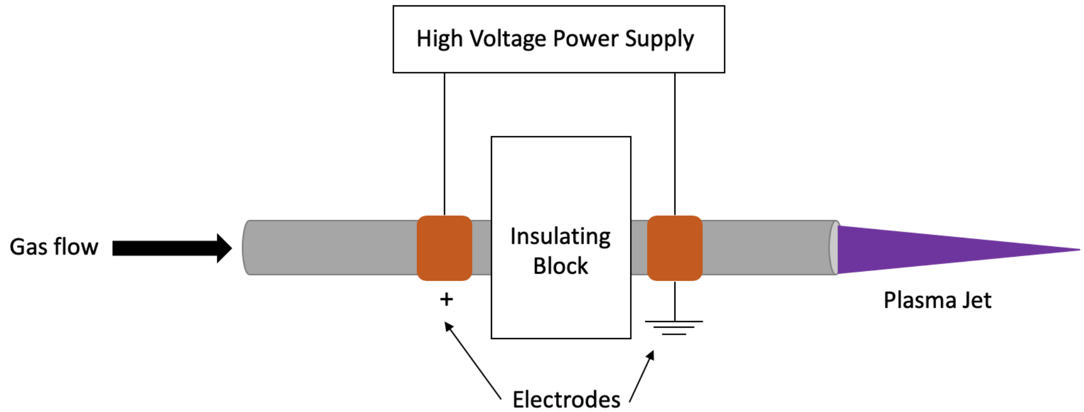

Figure 1: A typical ring to ring APPJ setup. The electric field forming the plasma is forced within the tube, thus forming a DBD jet.

Another common APPJ geometry is the inner electrode to base electrode. This features a grounded electrode *under* a sample with a centered inner electrode that extends within the length of a quartz tube housing flowing gas. This is well depicted by Winter [2]. An example setup is shown in Figure 2.

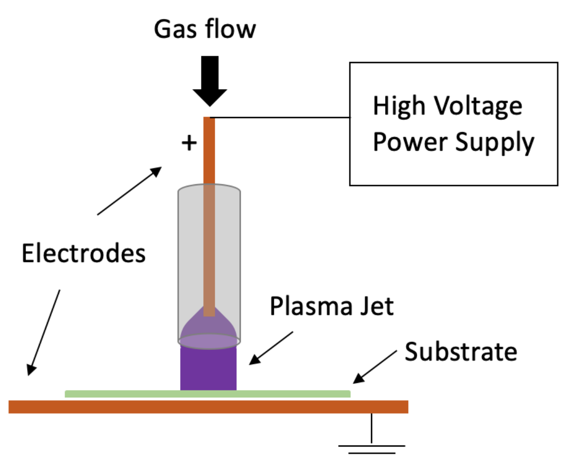

Figure 2: A typical inner electrode to base setup. In a way, the substrate itself (generally glass), acts as a dielectric for the DBD jet.

One final setup is the inner electrode to ring. As described by Chang [3], this setup requires the same centered inner electrode as above but features a grounded ring electrode outside the quartz tube. A basic inner electrode to ring setup is shown in Figure 3.

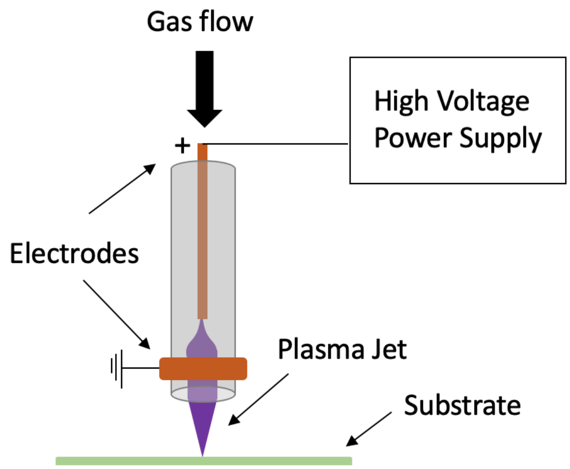

Figure 3: A typical inner electrode to ring setup.

For additional geometries, please consult Lu's work [4], which performs an extensive evaluation of other geometries not discussed here.

Final Design

It was determined that the inner electrode to ring setup best aligned with the needs of this project. For one, we already had the tools to easily create this setup from a previous experiment and had a tungsten inner electrode to use. Also, in the third setup, the two electrodes are only separated by one layer of quartz glass, compared to two layers in the first setup and the base electrode at a far distance in the second. From Paschen's Law, we find that the voltage required for plasma breakdown is dependent upon distance [5]. Adding additional dielectric material or elongating distance requires a higher voltage to be applied for breakdown. With the third setup, we could keep our voltages relatively low and within the operating regions of our equipment. With this setup determined as the most straightforward way to create a plasma jet, other, more long-term aspects of the experiment came under consideration. For one, it would be helpful to set up an Infrared Radiation (IR) camera to measure the temperature dispersion of a sample undergoing sintering. By adding a small mount to the setup, a FLIR Thermal Imaging Camera was mounted for future temperature observations. Another concern would be translating the sample under the jet in the x-y plane. Because we are not relying on a large base electrode to be placed under the sample, that area is free for a simple x-y translation stage to be placed under the jet. Although not included in the current design, this stage is something that could easily be implemented in the future. Also, it was decided that argon be used in this experiment due to its low breakdown voltage and inexpensiveness. After much trial and research into the subject, the physical and electrical parameters to create an acceptable jet were determined. The final setup, including electrical circuit, is shown in Figure 4.

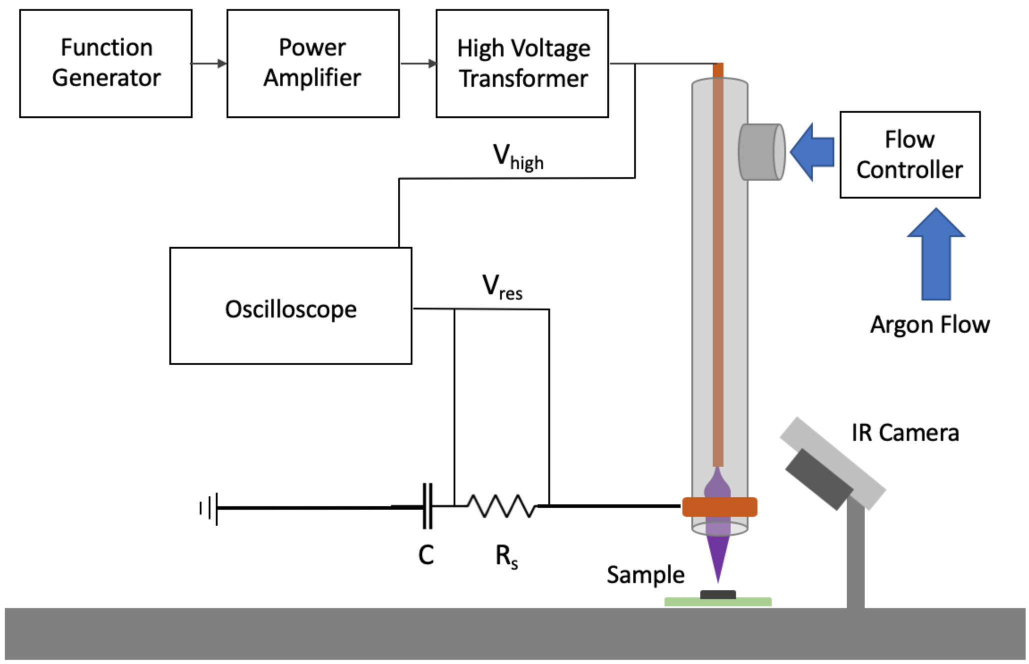

Figure 4: The final setup for this experiment. A stainless steel internal electrode was used while copper tape was wrapped around the tube exterior for the ring electrode.

For all proceeding experiments, a resistor with $R_s = 10$ kΩ rated to 3 W, and a capacitor of $C = 3$ nF were used. Argon was flowed at 20 slpm. Voltages ranging from 3 to 6 kV were applied at frequencies of 25 and 50 kHz.

Sintering Attempts:

Upon experimentation with the plasma jet, certain qualitative characterizations became clear. For one, increased voltage resulted in a thicker, brighter jet. An increase of frequency did the same. One can see in Figure 6 that for a higher input frequency, the internal column becomes noticeably wider.

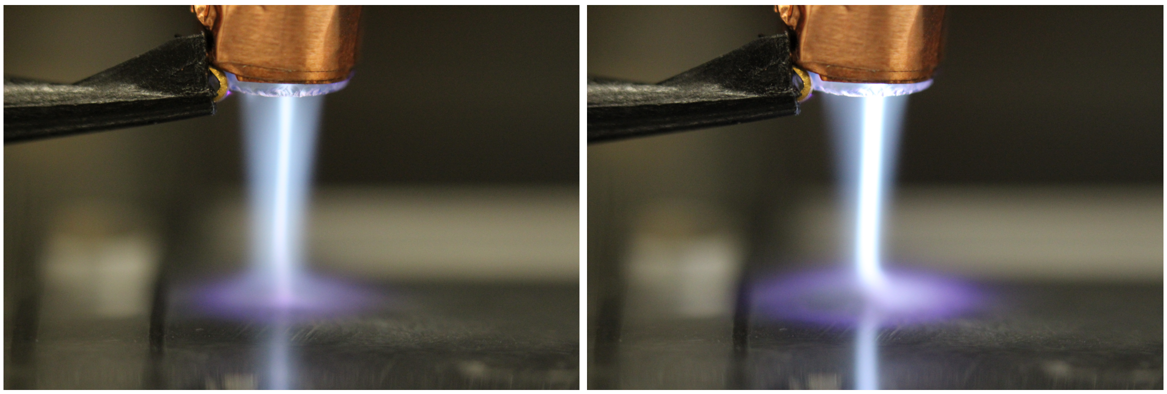

Figure 5: Argon APPJ at 25 kHz (left) and 75 kHz (right). One can clearly notice column thickness increases with frequency.

Although the main emphasis of this project was to create an adequate APPJ design, a small amount of sintering was also attempted on graphene and silver samples. 10 μm graphene samples of a 1×10 mm area were first experimented on. The qualitative result of a 120 s sintering attempt at 3.5 kV, 25 kHz, and 20 slpm flow is shown in Figure 6.

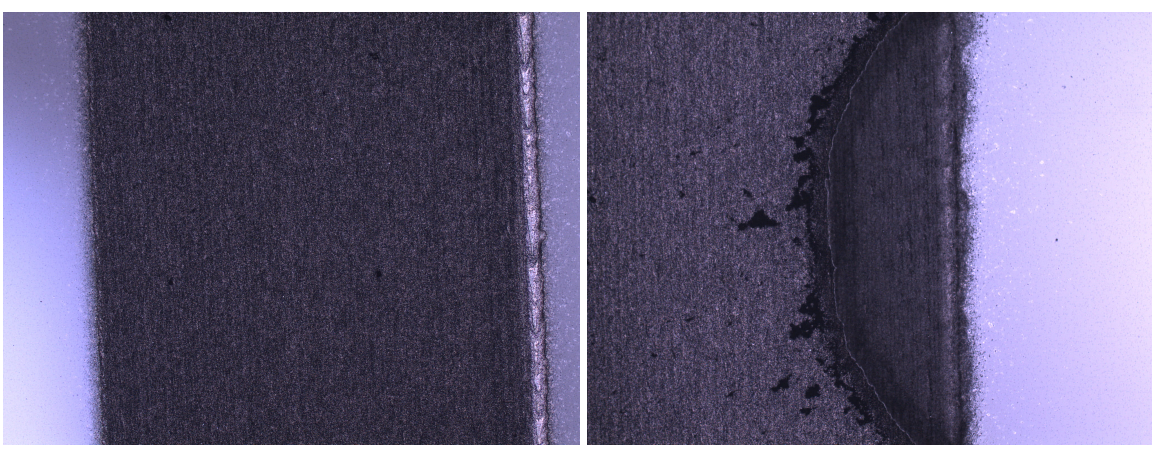

Figure 6: A 10× magnification of the sample before (left) and after (right) the 120 s exposure.

One can clearly see that the area where the central jet column had impacted was burnt up. This could potentially be averted by widening the plasma as was done in Figure 5. This, combined with x-y translation, could help spread the applied energy across the sample more evenly.

Other graphene experiments were performed where the central column of the jet was offset from the sample, allowing the sample to be exposed to a surface plasma instead. The resistance of the sample was measured before and after various periods of exposure. However, these results were relatively inconclusive. For instance, over a 220 s exposure time, the measured resistance dropped from 40 kΩ to 33.4 kΩ. However, upon re-measuring this sample the next day, the resistance had returned to 42 kΩ. This is likely due to the fact that the sample was measured directly after exposure, at a different temperature than steady state. Because graphene conducts heat so well, the resistance simply changed because the temperature changed, not because it was sintered. We retried the test, this time waiting 3 min after exposure. It was found that over exposure intervals of 10, 30, 60, and 120 s the resistance did not change at all. We also tested on silver, since silver is more robust, resistance is easier to measure, and it produces a visible shine when sintered. Also, sintered silver produces a resistance in the tens to hundreds of Ohms. Although there was not enough time to produce verifiable results, initial tests showed some promise. A 1 x 10 mm silver sample was exposed to the APPJ at 5.8 kV, 50 kHz, and 20 slpm argon flow for varying times. Three min after each exposure, the resistance was measured. The results for this preliminary experiment are shown in Figure 7.

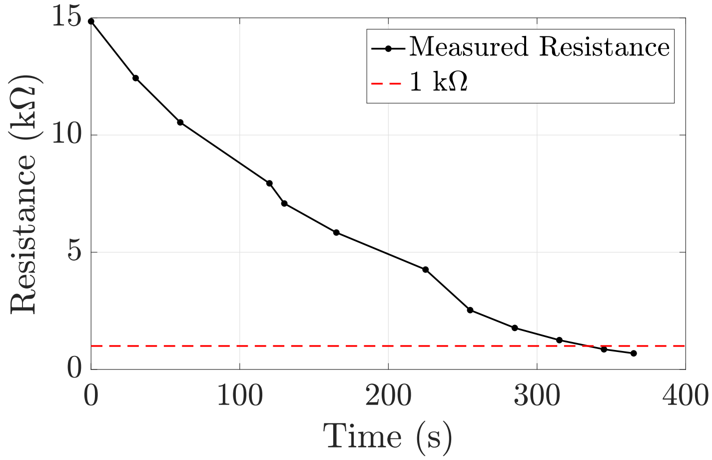

Figure 7: One can notice a clear trend of decreasing resistance with exposure. Remarkably, the silver sample which initially showed a 15 kΩ resistance was decreased 95% to 0.7 kΩ.

Discussion:

By showing a significant decrease in resistance, the silver sample became much more conductive. Clearly, exposure to the APPJ had a beneficial effect. This shows that, perhaps with renewed effort and some additional changes, this sintering method can be effective. For one, it would be beneficial to install the IR camera underneath the sample. With the sample being placed on a very thin substrate with a small Biot number, the temperature distribution of the sample should be accurately reflected on the bottom. By placing the IR camera directly underneath, a perfectly direct view of the distribution can be attained. Also, be implementing this in conjunction with a movable stage, the sample can be translated in the x and y directions, which will result in a much more evenly applied sinter. Additionally, the effect of varying flow rates and frequencies can be closely studied. Hopefully, in the future this project can be fleshed out to the point where an industrial process can be implemented using this technology. Ideally, this could save time and money versus conventional techniques, and lead to the continued advancement of materials science.

References:

[1] E A J Bartis *et al* 2013 *J. Phys. D: Appl. Phys.* 46 312002
[2] J Winter *et al* 2015 *Plasma Sources Sci. Technol.* 24 064001
[3] H Chang *et al* 2013 *Journal of Power Sources* 234 16-22
[4] X Lu *et al* 2012 *Plasma Sources Science and Technology* 21 034005
[5] Wikipedia contributors. “Paschen’s law.” *Wikipedia, The Free Encyclopedia*, 6 Oct. 2018.