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A new strategy of using dielectric barrier discharge plasma in tubular geometry for surface coating and extension to biomedical application

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ABSTRACT

There has always been a quest for nanotechnology to develop inexpensive coating methods with the capability of depositing biocompatible nanomaterials on biomedical and surgical tools. In this mini-report, a plasma-based innovative idea of coating a solid surface with antibacterial/antimicrobial nanosilver is floated and experimentally realized. The desired antibacterial nanosilver was obtained from laser ablation and directly entrained in an outflowing plasma jet, excited in the flow of argon at 10 l min−1 using 20 kV/20 kHz. Under these conditions, the jet can protrude 15 mm deeply into ambient air. The quality of the surface coating can be described by sparsely distributed particles or densely agglomerated clusters, controlled by the plasma length and the surface separation. Apart from the coating, plasma interaction leads to the sterilization of the exposed surface. The idea is essentially important to extend and upscale for coating biomedical and surgical devices in a flexible open processing environment.

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I. INTRODUCTION

Biomedical/surgical tools and transplants are extensively used in health centers world-wide. These are imperative in several medical fields including dentistry, neurology, cardiology, orthopedics, ophthalmology, and pharmacy. Surgical tools, such as retractors, clamps, sutures, gloves, and imaging devices, provide assistance to the surgeon in various processes during a surgery. Coating these surgical tools and medical devices with antibacterial/antimicrobial materials is one of the paramount needs at present. Nanotechnology has provided several possible solutions in this direction by projecting innovative nanofabrication ideas. While working in this capacity, recently, we were able to experimentally demonstrate a cold plasma method of coating antibacterial silver on a solid surface. The coating scheme basically employs a plasma jet from a dielectric barrier discharge (DBD) plasma device to transport the nanomaterial for deposition. The nanomaterial for deposition is produced by laser ablation and directly entrained in an outflowing plasma jet, excited in the flow of argon at 10 l min−1 using 20 kV/20 kHz. Under these conditions, the jet can protrude 15 mm deeply into ambient air. The quality of the surface coating can be described by sparsely distributed particles or densely agglomerated clusters, controlled by the plasma length and the surface separation. Apart from the coating, plasma interaction leads to the sterilization of the exposed surface. The idea is essentially important to extend and upscale for coating biomedical and surgical devices in a flexible open processing environment.
by electron charge density \( n_e \) and temperature \( T_e \), \( 10^9 \text{ cm}^{-3} - 10^{11} \text{ cm}^{-3} \) and 0.2 eV–5 eV, respectively. Furthermore, for these discharges, the plasma streamers, also called the plasma bullets, propagate at \( 10^4 \text{ ms}^{-1} - 10^6 \text{ ms}^{-1} \), several orders of magnitude higher than the gas flow, which clearly indicates that these are electrically driven rather than by the gas flow. To strike such a discharge, a sufficiently high AC voltage typically in the kV range is required but with a very small discharge current (\( \mu \text{A} \) to mA), thus making the power considerably insignificant. DBDs can be produced in different configurations; however, the double electrode configuration with the tubular geometry provides a more convenient and effective way to launch a plasma jet in the open air, essentially important for material processing and biomedical applications. Such plasma jets are easily adaptable to complex devices and conventional processes due to their easy operation in an open environment and due to their capability of launching plasma jets outside from the closed plasma system. In pharmacy, DBD plasma jets are employed to enhance the flowability of fine-grained powder to prevent the clogging of apparatus, precious product losses, and to sustain longer maintenance times. Further applications of an argon or helium plasma jet include wound healing, sterilization, and fabrication of nanostructures. Recently, Khan et al. applied this method for nanoparticle films of plasmonic materials for practical application in surface-enhanced Raman spectroscopy (SERS) for chemical detection. Similarly, in Refs. 14–16, some brief overviews have been given, describing the utility and motivation of the method for practical applications. The coating of biomedical tools with the desired material could be an interesting and novel application of DBD plasma jets with supplementary benefit of plasma sterilization. This would greatly facilitate the biomedical technology by treating large surgical tools and objects in the increased working space outside the main discharge regime.

In this mini-report, the idea of coating a solid surface with antibacterial nanosilver by an atmospheric argon plasma jet is presented. The antibacterial nanomaterial was obtained by the process of laser ablation at atmospheric pressure and entrained with an argon plasma jet for deposition on a located solid surface in open ambient. The nanomaterial landed on the surface is strictly adhered and formed a solid coating. The interaction of the plasma jet with the surface also provides some degree of sterilization. The aim of this study is to convey this idea of using a clean plasma source to gadgets for coating surgical tools and to upscale for industrial biomedical application.

II. EXPERIMENTAL PROCEDURE OF PLASMA EXCITATION

The experimental scheme of the atmospheric plasma coating strategy using an argon or helium plasma jet in the tubular geometry is shown in Fig. 1. In the given double electrode configuration, both the electrodes were placed outside the main discharge regime and wrapped around the gas feed tube of inner diameter 2.4 mm with a separation of 20 mm. The upstream electrode was driven by a high voltage AC of amplitude 15 kV–20 kV/20 kHz, and the downstream electrode was grounded. Placing the ring-shaped metal electrodes outside the tube constitutes a clean plasma source, as the plasma...
and the electrodes have no direct interaction, which is essential for the medical applications. Due to high AC voltage, a small discharge current flows through the gas, and a visible plasma is formed inside the tube while a plasma jet ejaculates outside the tube as a luminescence streamer as shown in the inset of Fig. 1. The plasma jet to target clearance was about 2 mm–3 mm to avoid the jet disturbance caused by its electrical nature with a metal surface. The plasma jet was used as a carrier of the coating material to the surface, exposed to the jet. The length of the plasma jet can be controlled by the gas flow, keeping all the other parameters fixed. The plasma is not only used as a coating streamer but also as a sterilizer for the coated surface, which is an added advantage. With this novel cold plasma approach, there are two possibilities: (1) the coating material of the pure target, produced by laser ablation, is directly transported with the plasma jet or (2) the coating material has to feed at the gas inlet to pass through the plasma active region and subsequently downstream in the plasma jet onto the surface. In both cases, the coating of the surface could be significantly different. We have used this approach to simply coat the antibacterial silver nanomaterial on a quartz, and it is described in this mini-report. The plasma was monitored with an iCCD camera, and the applied AC voltage was measured through a high voltage probe, attached to the live electrode. For the discharge current, a 47 Ω resistor was placed on the ground side of the electrode. The current–voltage signals were monitored on a digital oscilloscope not shown here.

III. RESULTS AND DISCUSSION

At first, to understand the operation mode of the plasma device used in the experiment, the voltage–current signals were measured through a digital oscilloscope. Figure 2 shows the typical voltage–current waveforms of the DBD plasma, operated in argon, supplied through a quartz tube at 10 l min$^{-1}$. During the positive half cycle of the applied AC voltage, a single current burst appears, which is repeated over the subsequent cycles. The peak amplitude of AC voltage and the discharge current were 20 kV/20 Hz and 55 mA, respectively. The current pulses are separated by 65 $\mu$s, which is considerably small, and the plasma apparently appears as continuous.
to a naked eye. For the electrical discharge, a visible argon plasma forms inside the tube called the plasma column with dimensions: length 28 mm and width 2.54 mm, and at the same time, a luminosity region, the so-called plasma jet, also appears, extending beyond the tube orifice as displayed in Fig. 2(b). For the operation conditions, the jet length and thickness were about 16 mm and <1 mm, respectively. The thickness of the jet seems uniform over the entire length. The current peaks that appeared in the waveform showed that the plasma device operates in the diffused mode, though some multiple smaller current traces are also observed. The diffused mode operation of the plasma is also well evident from the iCCD optical image displayed in Fig. 2(b). This behavior is important for biomedical application; however, the plasma can also be applied in the filamentary mode not shown here.

The formation of coating with the plasma is described in Fig. 3. A 104 × 140 μm² region of the scanned surface, coated with a silver nanomaterial using an argon plasma jet, is shown in Fig. 3(a). The coating nanomaterial was obtained from the process of laser ablation, which is not described here and can be read elsewhere (Refs. 10–12). The coating is particulate in nature and spreads over a region of about 2 cm–3 cm, visible to a naked eye. The particle mean size in the coating was about 28 (16) nm, where the number in the bracket represents the standard deviation in the particle size. The particle mean size was obtained using the lognormal function, fitted to the distribution of the Feret diameters of the particles obtained by ImageJ. Figure 3(b) shows the averaged line profile for the surface shown in (a). The profile shows fine surface features in the coating with a small variation across the scanned region. The surface plasmon resonance (SPR) shown in Fig. 3(d) was measured on the region, away from the plasma interaction zone, and it strongly confirms the plasmonic functionality and particulate nature of the coated film. The long tail of SPR, stretching in the infrared region of the absorbance spectrum, indicates an interactive behavior due to the dipole–dipole interaction of the particles in the produced coating. This character of particulate coating is important for several applications. In this strategy, both argon and helium could be equally important to use; however, the coating properties in both cases are significantly distinguishable as described in Ref. 11. The different coatings obtained in the two cases arise due to the different interaction behavior of these plasma jets and are not described here.

It is worth mentioning that to increase the deposition area in the case of coating large objects, a translation motion is required to feed into the surface using a translational stage. The surface is scanned and moves around the plasma jet, and a uniform coating is expected to obtain over a large surface area. The proposed nonthermal plasma coating strategy could be tried in the lab with an aim to apply and investigate for various types of coatings of surgical tools and to discern its feasibility for industrial scale biomedical application. At present, the work is new and passing through an initial stage of understanding; however, we expect sooner the method will appear as a promising clean plasma technique in the coating technology for biomedical devices and surgical tools.

IV. CONCLUSION

In conclusion, the nonthermal plasma coating methodology of antibacterial nanosilver was proposed and demonstrated in the lab. Our initial results suggested that DBD plasma jets seem useful to use as clean plasma streams for surface coating, certainly extendible to surgical tools and other biomedical devices. This method has the advantage of producing coatings of almost all materials along with the benefit of plasma sterilization, essentially required in the coating of surgical devices. Both argon and helium plasma jets could be equally effective in this strategy; however, the coating properties are significantly distinguishable in both cases. The length of the plasma jet is imperative and is controlled by the gas flow: shorter and longer jets are obtained at 10 at 4 l min⁻¹, respectively. Further research is encouraged to promote and develop this clean plasma coating methodology for a large scale biomedical application.

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DATA AVAILABILITY

It is to mention here that the design and operation of the DBD plasma setup along with the APLD setup could be provided upon request. The data that support the findings of this study are available from the corresponding author upon reasonable request.

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