Plasma Medicine, 8(1):45–55 (2018)

Helicon Gas Discharge for MUV-NUV Light Production

Alexander Hyde* & Oleg Batishchev

Department of Physics, Northeastern University, Boston, MA 02115, USA

*Address all correspondence to: Alexander Hyde, Physics Department, Northeastern University, 110 Forsyth Street, Boston MA, 02115, E-mail: a.hyde@neu.edu

ABSTRACT: A compact kW-level plasma source is being developed for EUV-UVA radiation and active radical production in potential biomedical applications under room conditions. Here we examine the MUV-NUV (200–400 nm) spectral range. The required strong B~0.1 T magnetic field domain is created using a chassis of rare-earth permanent magnets, bypassing the notorious issue of electromagnet overheating. Stable RF-driven discharge is achieved for a range of air pressures. The emission spectra are compared to that of high-purity nitrogen gas. Possible tuning of the spectrum, benefits of the helicon plasma, and further improvements of the source to maximize UV output are discussed.

KEY WORDS: helicon discharge, UV light source

I. INTRODUCTION

For several decades, there has been interest in the germicidal properties of plasmas. It has been shown that exposure to neutral and charged species contained in an ionized gas, as well as its ultraviolet (UV) light emission, can result in the deactivation or death of microorganisms.[1–5] The relative importance of each of these components de pends on a wide range of parameters[6,7] and has been the topic of intensive investigation. Many studies have focused on the etching effect of direct or indirect exposure to reactive atomic and molecular species.[1,8,9]

However, light in the UV range (100–400 nm) is well known as an effective standalone antimicrobial tool. It can be remotely applied and, much like direct nonthermal plasma treatments, is preferable to thermal sterilization when the target is heat sensitive.[10] As such, UV light has found a niche in the sterilization of medical equipment / surfaces,[11] air,[12] water,[13] and food.[14] The biological processes at work in such applications are complex, but generally they can be attributed to two separate mechanisms: direct absorption of the UV light by the DNA of the contaminants, leading to lesions that prevent replication, and erosion of the contaminants via photodesorption, wherein chemical bonds in the surrounding biological material are broken and the resulting reactive molecules damage individual cells of the sample.[4,15]

It is likely that multiple alternating phases of these two mechanisms produce the germicidal effects that have been observed.[16] These processes depend strongly on the wavelength of the ultraviolet light. Light in the UVC range (100–280 nm) is generally accepted to be the most efficient for germicidal purposes,[6,17] as the maximum in the

DNA and RNA absorption spectrum at 260 nm[12] lies in the spectral range below 275 nm where photons possess sufficient energy to break C–C and C–H chemical bonds.[10] UVB (280–320 nm) can be used for similar purposes with slightly less effectiveness.[2,14] However, because the absorption curve of DNA falls off exponentially above 320 nm,[3] UVA emission (320–400 nm) interacts less efficiently with DNA by several orders of magnitude.[14] This shortcoming can be addressed with larger dosage or through enhancement of the germicidal effect through the introduction of photosensitizers to the target.[14] UVA can also sterilize by other mechanisms like deteriorating cell membranes, damaging proteins, and producing harmful radicals,[13,14] while penetrating deeper into water[14] and posing less of a health risk to humans.[18]

It is by virtue of these applications that a need for strong, portable sources of UV light has arisen in the field of plasma medicine. Traditionally, monochromatic low-pressure lamps utilizing the ~254-nm mercury line or polychromatic medium-pressure versions producing a wide range of germicidal lines have been used.[19,20] These sources are not ideal from a practical perspective, as commercial models often take time to warm up before each use and their contents are toxic and must be disposed of after the fragile envelope containing it breaks or the relatively short lifetime of the device ends.[13] Furthermore, it is suspected that the sterilization resulting from such lamps takes place predominantly in the surface layer through direct absorption of the light,[16] and a concerns about their effectiveness have been raised.[21] As a result of these limitations, a variety of more robust plasma sources[8–10,22] have been developed for a wide range of medical purposes.[6,9]

While many types of gas discharge have been used in these sources, one underutilized option is helicon discharge.[23] It is maintained via the excitation of an electromagnetic mode, meaning that it requires a background magnetic field to propagate but necessitates no plasma-facing electrodes, which often degrade over the lifetime of such devices. Helicon plasmas also demonstrate efficient ionization, resulting in very high, continuous plasma densities up to 10^{20} cm^{-3}.[24] They are also typically ignited at low ambient pressures, which necessitates sophisticated vacuum equipment that may be impractical in a medical setting.[25] We have, however, identified a promising operational regime between ~10^{-1} and 10 torr.[26] This pressure range can be achieved with relatively simple equipment while avoiding altogether the issues with discharge ignition at higher pressures. We have ignited radio frequency (RF) discharges in this intermediate pressure regime with arrays of permanent magnets providing the magnetic field. Preliminary results suggest that the discharge can serve as an effective source of UV light, EUV light, and radicals.[27]

In this paper, we present an in-air helicon discharge based on our previous work but redesigned to serve as a source of UV light for biomedical applications. The setup consists of a permanent magnet chassis and an RF antenna that ionizes gas inside a fused-silica tube. The pressure is maintained in the desired operational range via a portable mechanical vacuum pump. This compact, closed system can generate light emission across the UVA–UVC range. To demonstrate this capability, spectra of the emission from several different gas discharges are presented. These results are analyzed and discussed, and possible improvements to the design are considered.

II. EXPERIMENTAL SETUP

The experimental system used to generate light emission for these tests is an extension of our previous work with the minihelicon plasma source.[28] A schematic of the layout is given in Fig. 1. It utilizes a helical antenna with circular cross-section, immersed in a domain of solenoid-like magnetic field, to ignite the helicon mode in a quartz tube. Neutral gas is supplied and regulated from one end of the tube and pumped out through the other. As such, the system is closed and the discharge is isolated from the environment. The only byproduct of the plasma that escapes is its spectral emission. The quartz tube, transparent above ~180 nm, permits light in the UV–VIS–NIR range to propagate to the surrounding environment. This in-air design allows the targets to be brought close to the discharge, thereby maximizing UV light exposure while main taining them at standard room conditions.

The required magnetic field is produced with an array of permanent magnets. This avoids the need for external DC power that would be introduced by us ing an electromagnet, as is common. The magnets, cylindrical in shape and of N45 grade neodymium, are contained in a rigid aluminum chassis that takes the form of a hexagonal prism. They are secured in close-fitting holes that pattern the flat faces of the chassis in a hon-

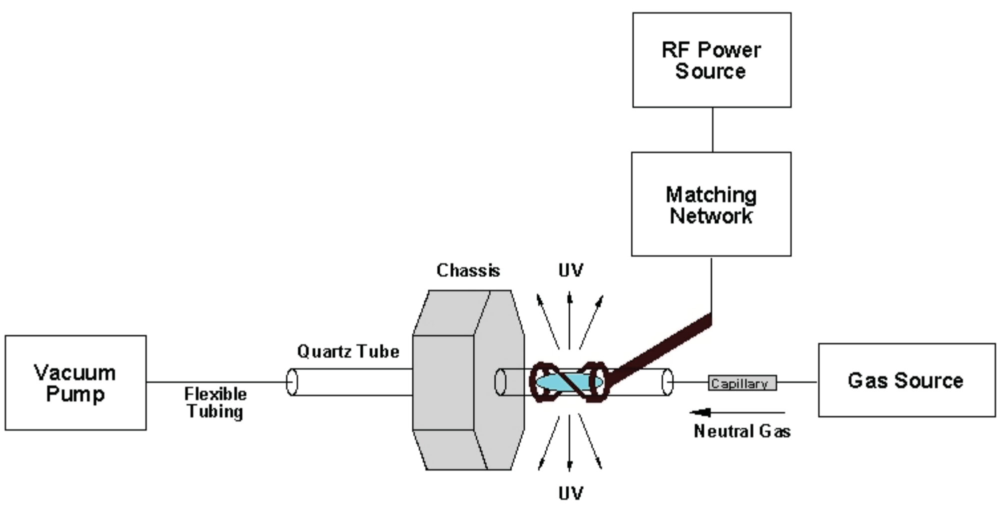

FIG. 1: Schematics of the experiment: gas is supplied into and excited inside the quartz tube by a helical antenna, powered by the 13.56 MHz RF-source. A hollow chassis of permanent magnets creates a strong axial magnetic field B~0.1 T in the antenna region.

eycomb arrangement. The field topology emitted by this chassis has been characterized extensively.[29] It consists of three strong field peaks: one in the bore of the chassis and two outside its faces on the central axis. A measured profile of the field along the central axis is given in Fig. 2. Each of the three field peaks can be used to support helicon discharges.[27] This arrangement is somewhat atypical, as the classical helicon mode requires a uniform field to propagate. For the purposes of these studies, one of the outer peak field regions are used.

To do so, the ~0.5 m long, double-spout quartz tube is arranged such that it rests in the central bore of the chassis and extends outwards on each side. The helical antenna is slid around the tube and positioned at the location of one of the outer field peaks. The antenna is connected to an RF generator via an impedance matching network. This allows the discharge to be toggled on and off nearly instantaneously. While up to 1.2 kW can be produced by the generator, we estimate that only ~50% of the applied power is delivered to the gas in the tube due to poor coupling and losses in the RF feeds.

Gas is supplied to the ionization region through flexible plastic tubing attached to the spout nearest the antenna. Flow is regulated using a capillary filament attached in series to this tubing. The desired pressure regime is obtained by pumping gas from the opposite side of the quartz tube with a portable mechanical pump. The pumping speed is ~200 L/min. In effect, this creates a neutral gas pressure gradient across the quartz tube, with the largest value at the gas inlet. The pump possesses a gauge that indicates the pressure at the pump outlet. However, there is a non-negligible length of plastic tubing between the pump and the rest of the instrumentation. As such, the exact pressures at points throughout the quartz tube are not precisely known. We report the outlet pressure *Po* for reference in this paper.

While the pressure can be increased above this range by flowing more gas into the

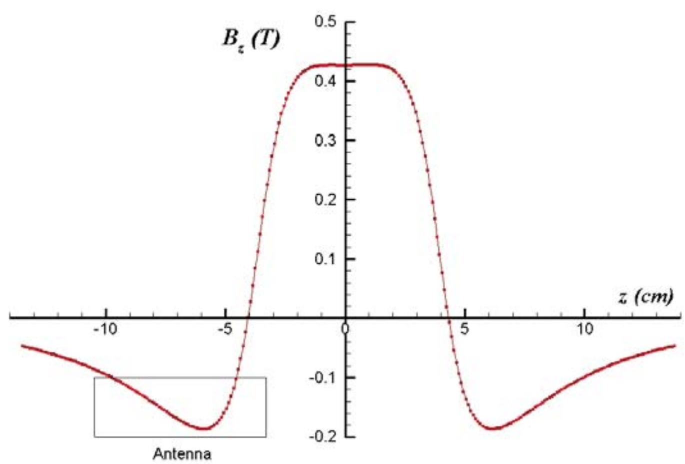

FIG. 2: Profile of the axial magnetic field component along the central axis of the magnet array. The rectangular box indicates the position of the helical antenna.

tube, the discharge quickly becomes very difficult to ignite. Higher pressure operation can be attained, however, by exploiting hysteresis in the system. If the source is ignited at low *Po* and the pressure is subsequently raised, the discharge will persist at pressure points far higher than could be otherwise achieved. This functionality opens the possibility of expand the range of operational parameters of the source.

III. RESULTS

For the purposes of these investigations, nitrogen gas with N4.5 purity and air were used. Discharges were ignited at a variety of pressures and power inputs. Emission spectra in the 200–900 nm UVVIS range were collected using an Ocean Optics USB2000 spectrometer. For added safety, the setup was mounted inside a vacuum facility equipped with RF feedthrough.

A. Nitrogen

Using the experimental setup described above, N_2 discharges could be directly ignited across a wide range of gas outlet pressures from 10^{-1} to ~300 mtorr. A photo of a typical plasma is contained in Fig. 3. Figure 4 shows the light emitted by such a discharge in the combined MUV and NUV spectral range at several different powers and pressures.

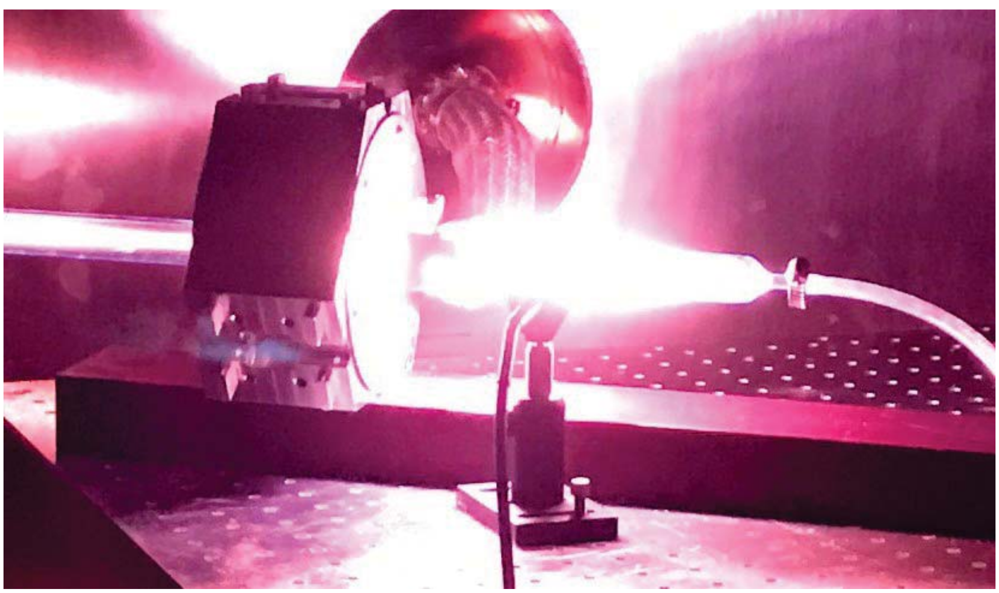

FIG. 3: A photo of helicon source operation on N_2 inside a vacuum tank vented to 1 atm. The primary components of the experiment are visible: the chassis, fused-silica tube, and the flexible tubing. The antenna is masked by the oversaturated light source. The RF feedthrough is visible in the back, while a fiber optics cable, attached to a collimator on a holder, is visible in front of the discharge.

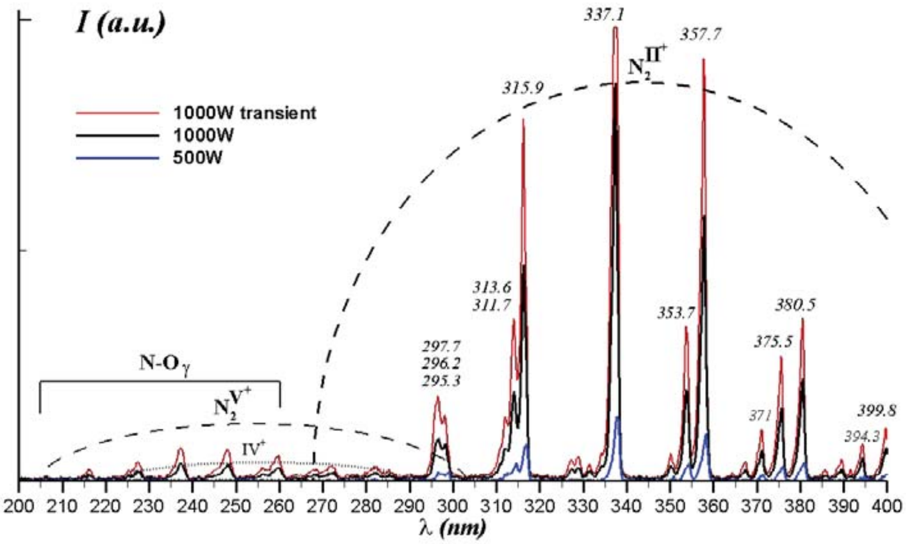

FIG. 4: Combined spectra of the N_2 helicon discharge for 3 different cases with varied RF power and initial conditions. The majority of the UVA emission derives from the second positive system of molecular nitrogen. One can see the non-linear UVA output vs input RF power. In the higher power density case, the spectrum is shifted towards the lower wavelengths, and bands in the UVC region appear. Their intensity doubles in the transient regime for the same input power.

Despite the brightness in Fig. 3, the vast majority of line emission takes place in this 200–400-nm window. Strong emission from the band associated with the second positive system of N_2[30] is visible throughout the 270–400-nm range. A series of unidentified band heads are also visible between 200 and 300 nm. These could be associated with the fifth positive system of N_2[30] or with the nitric oxide gamma band N-O$_\gamma$ commonly cited in the literature[4,5] for gas discharges with similar composition. At present, the data are insufficiently precise to distinguish between these two possibilities. The majority of the VIS emission is accounted for by weak excitation of the first positive system of N_2.

Generally, the intensity of the light emission grows as more power is coupled to the gas. However, our observations indicate that this relationship is not strictly linear. Instead, the discharge appears to undergo an abrupt change in brightness, and possibly composition, at some point while the power is being gradually increased. We suspect that this represents a transition between inductively coupled and helicon modes in the plasma, a well-documented effect for such discharges.[24,28] The 500 and 1000 W discharge data in Fig. 4, ignited at P_o ~300 mtorr, demonstrate these dim and bright modes, respectively.

The bright helicon mode was significantly easier to obtain at lower pressures. However, by initiating the discharge in the $P_o = 10^{-1}$–10 mtorr range and then gradually increasing the pressure, it could be sustained up to P_o ~300 mtorr without immediate reversion to the dimmer mode. Emission data collected in this transient regime, as the pressure is gradually being increased, are included in Fig. 4. The resulting spectral lines are more intense than those produced when the discharge is ignited at P_o = 300 mtorr directly. Comparison of the relative intensities of individual spectral lines indicates that the magnitude of this effect varies according to wavelength. The 371-, 375.5-, and 380.5 -nm heads of the second positive system, for instance, increase in intensity by 50%. However, emission from the unidentified band in the MUV region doubles. Overall, the prevailing trend appears to be that when further energy is deposited in the gas, it is radiated disproportionately from lower wavelength, higher energy UV lines. This can be attributed to higher electron temperature *Te*, resulting from better RF coupling to the plasma.

B. Air

Helicon discharges are easily achievable in air as well. Figures 5 and 6 display the helicon air discharge and its associated spectrum, respectively. As in the nitrogen discharge, a large portion of the emitted NUV light arises from excitation of N_2's second positive system. Unlike the previous case, however, there is also strong emission between 307 and 311 nm from a band associated with the hydroxyl radical OH.[31] Excitation of the unidentified band in the MUV region is minimal in this case. By color, the air discharge in Fig. 5 is visibly whiter than the N_2 discharge pictured in Fig. 3. This is due to the pres-

FIG. 5: A photo of helicon source operation on air taken through a viewport of the vacuum tank. The color is whiter, and the intensity is higher compared to the pure N_2 shown in Fig. 3.

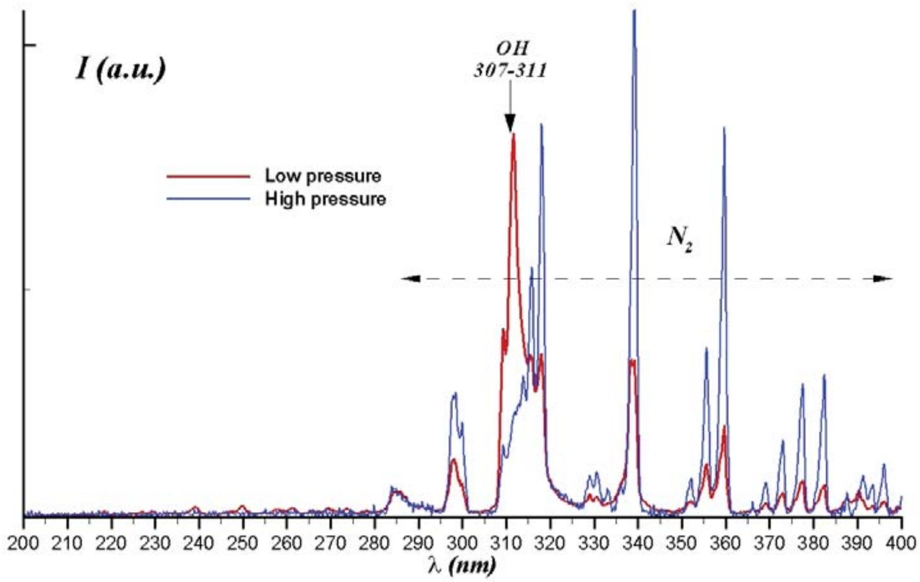

FIG. 6: Spectra of an air helicon discharge for two extreme cases. At high pressure, the UVA region is dominated by the same N_2 bands as in Fig. 4, while the UVC spectrum is barren. At low pressure, the UVB band associated with hydroxyl OH emission is strongest and the UVA region is depleted, but the same UVC bands (see Fig. 4) protrude again.

ence of the hydrogen Balmer series and atomic oxygen lines superimposed on top of the band from N_2's first positive system.

Spectroscopic data collected at two different P_o's (10^{-1} and ~300 mtorr) are presented in Fig. 6. In the low-pressure case, the hydroxyl band dominates and the nitrogen band is deemphasized. At higher pressures, the situation is reversed. This may be a reflection of a fundamental difference between the modes propagating in the two discharges or of differing compositions of the gases in each. The source of the OH radical is not immediately apparent, and as such, its prevalence in the air used is unknown.

IV. DISCUSSION

The results presented above suggest that the proposed setup holds promise as a source of near and middle ultraviolet light. The N_2 discharge in particular emits the majority of its light in this range, and exhibits significant emission in the 200–270-nm germicidal range. To determine the source of this radiation, the unidentified UVC band observed there will be investigated with a higher-resolution spectrometer. If it is indeed the $N–O_\gamma$ band, it is possible that the oxygen contained therein originates in the small 30 ppm impurity present with the N_2 gas.

The apparent dependence of the UV emission on the initial pressure conditions at which the dischargeis ignited is of further interest. The presence of this effect suggests that the ratio of UVC to UVA+B light can be scaled through T_e according to which is pre ferred for a given application. In the case of the air discharge, the origin of the hydroxyl radical must be identified to determine what fraction of the air mixture it accounts for. For the N_2 discharge, it should be determined whether the increased electron temperature depends on the initial conditions at which the discharge is ignited or simply on RF power density.

Beyond the changes it induces in the emission spectrum, the ability to ignite and then gradually raise the gas pressure permits the use of simple vacuum equipment. In the future, however, reliance on such equipment should be eliminated altogether. Furthermore, while the outlet reading given by the pump serves as a good indicator of changes in the pressure, the gas pressure profile in the tube is unknown. Thus, in the short term, precise measurement of the pressure in the tube is required.

Another outstanding issue remains that while the source's MUV–NUV–VIS emission has been documented here, its EUV–VUV spectrum has not been investigated (although 108–113 nm line emission has been observed for N_2 helicon plasma source operation in vacuum[27]). Light carrying these high energies plays a role in sterilization,[5,21] but the quartz tube is not transparent to such wavelengths. Measurement of the emission in this region is of interest. Future designs that incorporate a way to utilize this light, or gain access to the plasma and radicals we observe in the discharge, could exploit the germicidal properties of each of these outputs[4,8] as well as those of the synergies between them.[13,15,16]

V. CONCLUSIONS

A portable helicon experiment was built to demonstrate UVC–UVA light production under common room conditions for possible biomedical applications. An array of permanent magnets was used to create the high, quasi-uniform magnetic fields up to 0.5 T that are required to facilitate helicon mode excitation and gas ionization. Up to 1kW of RF power was deposited into the plasma, indicating efficient gas ionization and good electron confinement by the magnetic field.

For initial demonstration, atmospheric gases and air have been used. For high-purity nitrogen with varied input power, nonlinear response of the emitted UV light intensity to the input power was observed. The shift toward short wavelengths is attributed to increasing electron temperature. Discharge bifurcation, namely achieving different steady-state conditions for the same input power, was observed. In the transient pressure regime, UVC emission is doubled, which may be due to improved RF–plasma coupling. The spectroscopic system uti lized does not allow us to decipher this region, and higher spectral resolution is required. We determined that the majority of the UVA emission lines are created by the second positive system of molecular nitrogen, and VIS emission from the first positive system is observed, which makes the discharge pink in appearance in Fig. 3.

Air discharge demonstrates similar domination of nitrogen molecular bands from the first and second systems at higher pressures, but emission in the UVC region is almost nonexistent. With reduced pressure, the spectrum changes dramatically: a strong UVB hydroxyl emission band appears and eventually dominates the spectrum. At the same time, the Balmer series of hydrogen emerges in the VIS region, along with strong atomic oxygen lines. As a result, the discharge color changes from pink to white, with much increased overall intensity.

Future work and potential changes to the design were also discussed. Namely, our desire to measure the exact gas pressure in the source and further simplify the vacuum equipment are mentioned. Additional studies with higher resolution spectroscopy were suggested to identify the bands that produced the UVC emission. Deeper investigation into the source of the hydroxyl in the air spectra and the EUV-VUV emission currently blocked by the fused silica tube are also merited.

ACKNOWLEDGMENTS

The research work at NU Plasma Laboratory was performed with the support of US AFOSR Grant FA9550-10-1-0498 and US DoD Award FA2386-12-1-3006. We thank our colleague Mr. Timothy Hussey for help with the portable vacuum pump.

REFERENCES

1. Fridman G, Friedman G, Gutsol A, Shekhter AB, Vasilets VN, Fridman A. Applied plasma medicine. Plasma Proc Polymers. 2008; 5(6):503–533.
2. Lindberg C, Horneck G. Action spectra for survival and spore photoproduct formation of Bacillus subtilis irradiated with short-wavelength (200–300 nm) UV at atmospheric pressure and in vacuo. J Photochem Photobiol B: Biol. 1991;11(1):69–80.
3. Sutherland J, Griffin K. Absorption spectrum of DNA for wavelengths higher than 300 nm. Radiat Res. 1981;86(3):399–410.
4. Boudam MK, Moisan M, Saoudi B, Popovici C, Gherardi N, Massines F. Bacterial spore inactivation by atmospheric-pressure plasmas in the presence or absence of UV photons as obtained with the same gas mixture. J Physics D: Appl Physics. 2006;39(16):3494–507.
5. Brandenburg R, Lange H, von Woedtke T, Stieber M, Kindel E, Ehlbeck J, Weltmann KD. Antimicrobial effects of UV and VUV radiation of nonthermal plasma jets. IEEE Trans Plasma Sci. 2009;37(6):877–83.
6. Kong MG, Kroesen G, Morfill G, Nosenko T, Shimizu T, van Dijk J, Zimmermann JL. Plasma medicine: an introductory review. New J Physics. 2009;11(11):115012.
7. Rossi F, Kylian O, Rauscher H, Hasiwa M, Gilliland D. Low pressure plasma discharges for the sterilization and decontamination of surfaces. New J Physics. 2009;11(11):115017.
8. Laroussi M. Nonthermal decontamination of biological media by atmospheric-pressure plasmas: review, analysis, and prospects. IEEE Trans Plasma Sci. 2002;30(4):1409–15.
9. Ryan TP, Stalder KR, Woloszko J, Overview of plasma technology used in medicine. Proceedings of the SPIE, 8584, 2013 Feb 26.
10. Halfmann H, Denis B, Bibinov N, Wunderlich J,Awakowicz P. Identification of the most efficient-VUV/UV radiation for plasma based inactivation of Bacillus atrophaeus spores. J Physics D: Appl Physics. 2007; 49(19):5907–11.
11. Pollak J, Moisan M, Kroack D, Boudam MK. Low-temperature low-damage sterilization based on UV radiation through plasma immersion. J Physics D: Appl Physics. 2008;41(13):135212.

12. Green CF, Scarpino PV. The use of ultravioletgermicidal irradiation (UVGI) in disinfection of airborne bacteria. Environ Engineer Policy. 2001;3(1):101–7.
13. Chatterley C. Linden K. Demonstration and evaluation of germicidal UV-LEDs for point-of-use water-disinfection. J Water Health. 2010;8(3):479.
14. Bintsis T, Litopoulou-Tzanetaki E, Robinson RK. Existing and potential applications of ultraviolet light in the food industrya critical review. J Sci Food Agri. 2000;8(6):637–45.
15. Moisan M, Barbeau J, Crevier MC, Pelletier J, Philip N, Saoudi B. plasma sterilization methods and mechanisms. Pure Appl Chem. 2002;74(3):349–58.
16. Philip N, Saoudi B, Crevier MC, Moisan M, BarbeauJ, Pelletier J. The respective roles of UV photons and oxygen atoms in plasma sterilization at reduced gas pressure: the case of N_2O_2 mixtures. IEEE Trans Plasma Sci. 2002;30(4):1429–36.
17. Marshall MM, Hayes S, Moffett J, Sterling CR,Nicholson WL. Comparison of UV inactivation of spores of three encephalitozoon species with that of spores of two DNA repair-deficient Bacillus subtilis biodosimetry strains. Appl Environ Microbiol. 2003;69(1):683–5.
18. Mori M, Hamamoto A, Takahashi A, Nakano M, Wakikawa N, Tachibana S, Ikehara T, Nakaya Y, Akutagawa M, Kinouchi Y. Development of a new water sterilization device with a 365 nm UV-LED. Med Bio Engineer Comput. 2007;45(12):1237–41.
19. Chen RZ, Craik SA, Bolton JR. Comparison of the action spectra and relative DNA absorbance spectra of microorganisms: Information important for the determination of germicidal fluence (UV dose) in an ultraviolet disinfection of water. Water Res. 2009;43(2):5087–96.
20. Moisan M, Barbeau J, Moreau S, Pelletier J, Tabrizian M, Yahia, L. Low-temperature sterilization using gas plasmas: a review of the experiments and an analysis of the inactivation mechanisms. Int J Pharmaceut. 2001;226(1):1–21.
21. Lerouge S, Fozza AC, Wertheimer MR, Marchand R, Yahia L. Sterilization by low-pressure plasma: the role of vacuum-ultraviolet radiation. Plasma Polymer, 2000;5(1):31–46.
22. Awakowicz P, Baldus S, Stapelmann K, Engelhardt M, Bibinov N, Denis B. Optical emission spectroscopy as a tool for characterization of technical plasmas in medical applications. Plasma Med. 2012;2(1–3):151–68.
23. Chen F. Plasma ionization by helicon waves, Plasma Physics Control Fusion. 1991;33(4):339–64.
24. Ellingboe AR, Boswell RW. Capacitive, inductive and heliconwave modes of operation of a helicon plasma source. Physics Plasma. 1996;3(7):2797–804.
25. Laroussi M. Sterilization of contaminated matter with an atmospheric pressure plasma. IEEE Trans Plasma Sci. 1996;24(3):1188–91.
26. Hyde A, Kamieneski R, Batishchev O. Development of a compact atmospheric pressure plasma source. Bull APS. (Providence, RI). 2012;57(12): 324.
27. Hyde A, Taylor A, Batishchev O. Permanent magnet chassis for UV light and plasma sources. IEEE Trans Plasma Sci. In Press 2018.
28. Batishchev O. Minihelicon plasma thruster. IEEE Trans Plasma Sci. 2009;37(8):1563–71.
29. Hyde A, Bushmelov M, Batishchev O. Tunable system for production of mirror and cusp configurations using chassis of permanent magnets. J Magnet Magnetic Mater. 2018;449(1 March 2018):197–206.
30. Lofthus A, Krupenie PH. The spectrum of molecular nitrogen. J Phys Chem Ref Data. 1977;6(1):113–307.
31. Li Q, Takana H, Pu YK, Nishiyama H. A nonequilibrium argon-oxygen planar plasma jet using a half-confined dielectric barrier duct in ambient air. Appl Physics Lett. 2012;100(13):133501.