

Provided for non-commercial research and education use.
Not for reproduction, distribution or commercial use.



This article appeared in a journal published by Elsevier. The attached copy is furnished to the author for internal non-commercial research and education use, including for instruction at the authors institution and sharing with colleagues.

Other uses, including reproduction and distribution, or selling or licensing copies, or posting to personal, institutional or third party websites are prohibited.

In most cases authors are permitted to post their version of the article (e.g. in Word or Tex form) to their personal website or institutional repository. Authors requiring further information regarding Elsevier's archiving and manuscript policies are encouraged to visit:

<http://www.elsevier.com/copyright>



Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Surface treatment comparison using atmospheric helium plasma jets with different frequencies and target objects

Dan Bee Kim¹, B. Gweon, S.Y. Moon², W. Choe*

Department of Physics, Korea Advanced Institute of Science and Technology, 335 Gwahangno, Yuseong-gu, Daejeon 305-701, Republic of Korea

ARTICLE INFO

Available online 24 March 2010

Keywords:

Atmospheric pressure plasma
Corona
Frequency
Surface treatment

ABSTRACT

This paper details a plasma surface treatment feasibility study conducted for different driving frequencies of low frequency (LF, 50 kHz) and radio-frequency (RF, 13.56 MHz) sources by adding a target plate to a single pin electrode plasma jet system. The plasmas were generated in ambient air with a helium gas supply. The electrical characteristics of the plasmas were studied and compared for cases with and without a plane electrode. The LF plasma was more affected by the presence of the dielectric plate than was the RF plasma, showing a larger increase in its current. In addition, pork samples were treated to assess the electrical and thermal safety of plasma biomaterial surface treatments. The LF plasma conduction current and temperature during the pork treatment were 0.5 mA and 22 °C, and those of the RF plasma were 40 mA and 31 °C at maximum.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Atmospheric pressure plasmas have attracted much interest in the last several decades, and many research approaches have been applied to addressing plasma source geometry, generation power type, and diagnostics, etc. [1–8]. To complement or replace the existing low pressure plasmas employed in industry, especially in the manufacturing processes of semiconductors and displays, etc., generation of a large area or volume plasma with a high spatial uniformity was the first focus of research [1,9]. Recently, generation of small volume plasmas has been emphasized instead, as it can become a unique feature of the atmospheric pressure plasmas. Small size atmospheric pressure plasmas have their own merits of offering better stability, high electron energy, and high radical density, so they can be applied to various applications such as light sources, displays, gas detectors, and bio and medical treatments, etc. [10–13].

Plasma bio/medical applications using small size atmospheric pressure plasma have been particularly interesting to many researchers. Several small size jet type atmospheric pressure plasmas have been developed, and they have been used to treat many kinds of germs on culture grounds and also for the treatment of a few living cells. Most of these small plasmas sources have employed radio frequency (RF) [14,15] as the RF power supplies are rather common, although not necessarily better, and a few studies have employed low frequency (LF) [16] and microwave [17]. Although different source frequencies

induce distinct plasma features and, thus, certain frequency ranges may be more suitable than others, research into driving frequencies has not received adequate attention. A comparative study of various frequency ranges has just been undertaken; our group compared the characteristics of both LF and RF single pin electrode type plasmas [3], and Kim et al. attempted inactivation of *E. coli* with both LF and RF coaxial plasmas [18]. Further detailed study will be required to successfully and efficiently employ jet type plasmas. A close study of plasma characteristics is required for fundamental physics information and to help analyze and improve the application results. For instance, basic electrical measurement of the current flow through the application target has not been made for the jet type system.

In our previous work, small size jet type plasmas were generated in a single electrode system using two different input frequencies (50 kHz and 13.56 MHz). The plasmas exhibited distinct characteristics due to their frequency difference, but they commonly had low current and gas temperature properties with adequate features for bio/medical applications [3]. In this work, a plane target was added to the system in order to study the feasibility of using LF and RF plasmas for surface treatments and also to distinguish their characteristics. In addition, a prepared sample of pork, often used as a biological model for humans due to similarities in human and porcine size, physiology, organ development and disease progression, was treated with both plasmas to assess the electrical and temperature safety of the plasma treatment of living organisms [19,20].

2. Experimental setup

The single electrode plasma generation source used in this study consists of a copper pin electrode with a radius of 360 μm placed in a

* Corresponding author.

E-mail address: wchoe@kaist.ac.kr (W. Choe).¹ Present address; Korea Research Institute of Standards and Science.² Present address; LG Electronics Advanced Research Institute.

pyrex tube with a 3 mm radius. With the helium gas supplied through the pyrex tube at a gas flow rate of 3 slpm, plasmas were generated by either 50 kHz LF (FTLab HPSI200) or 13.56 MHz RF (YS E06F) power in the ambient air. Details of the generation source are reported elsewhere [3,7]. When a plane target was added and when the pork sample was treated, the inter-electrode distance was kept at 6 mm under most circumstances.

After the plasma generation, electrical diagnostics were performed using a Tektronix P6015A voltage probe and a Pearson 4100 current probe for the LF plasma and using a Prosys EI-100 voltage and current probe for the RF plasma. Gas temperature was measured as well using a FISO FOT-H fiber thermometer.

3. Results and discussion

3.1. Effects of the dielectric plane electrode

The single electrode system is favorable for its simple configuration, easier access to targets and portability. However, in real applications, when the target objects are placed in front of the plasma, the plasma characteristics may be changed by the presence of these objects. Considering that many target objects like polymers and biomaterials are dielectric, a dielectric plate was used as a target in this study.

Fig. 1 shows images of LF and RF plasmas. Without the plane target, the LF plasma appears cylindrical in shape and several tens of millimeters in length [Fig. 1(a)], while the RF plasma is spherical and only a few millimeters in length [Fig. 1(b)]. This difference in the physical appearance of the plasmas is due to the different driving frequencies. According to the electron displacement equation for an electron under a sinusoidal external electric field at a very frequent collisional condition, a smaller input frequency results in a larger electron displacement. As a result, the LF plasma is much longer than the RF plasma [3].

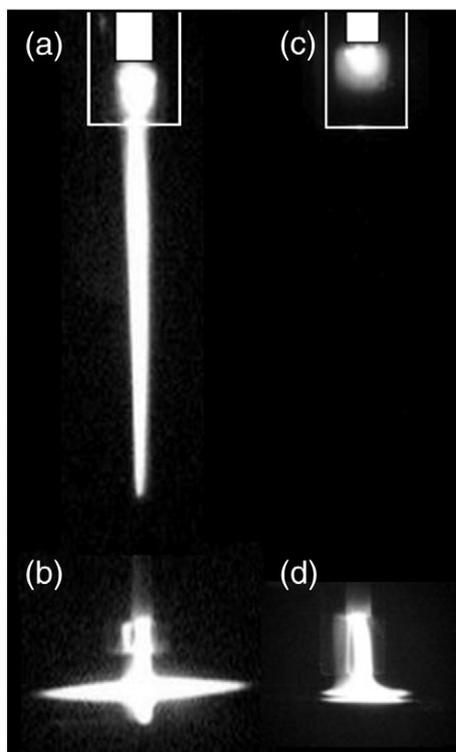


Fig. 1. Images of LF plasmas (a) without and (b) with the target plate and images of RF plasmas (c) without and (d) with the target plate.

With a conducting material coated at the backside of the dielectric plane [indium tin oxide (ITO) glass] as a target plate, both plasmas spread on it and form a cone-like shape due to the dielectric surface charging, as shown in Fig. 1(c) and (d). Unlike the LF plasma, which spread onto the dielectric ground electrode at the breakdown, the RF plasma was generated only near the powered pin electrode at the breakdown and at low input power. Also, the plasma area at the dielectric surface was larger for the LF plasma. Such differences can be explained by the different input frequencies, as it is difficult to detect dielectric charging at the dielectric surface at RF [21].

The discharge voltage and current are plotted in Fig. 2(a) for the LF plasma and in Fig. 2(b) for the RF plasma. The slope of the discharge current–voltage (I - V) characteristic curve indicates the impedance (Z) of the plasma from $V=ZI$ and $Z=R+i\omega L+1/i\omega C$, where R , L , and C denote the resistance, inductance and capacitance of the discharge, respectively. Looking at the plots, the impedance was decreased by 40% for the LF plasma and by 5% for the RF plasma by employing the ITO target plate. Usually, the plasma bulk is seen as R , and the sheath is seen as C connected in series. Then, the increase in the electron density results in the decrease in R of the plasma bulk and the increase in C of the plasma sheath [22]. As a result, the total plasma impedance decreases. Therefore, assuming that the reactance was changed little, the decrease in the resistance suggests a higher electron density or energy.

The conduction current was calculated by subtracting the displacement current from the total measured current. The LF plasma conduction current with the target plate was between 1.1 mA and 2.2 mA, while that without the plate was between 0.03 mA and 0.16 mA for the same discharge voltage range. The similarly calculated

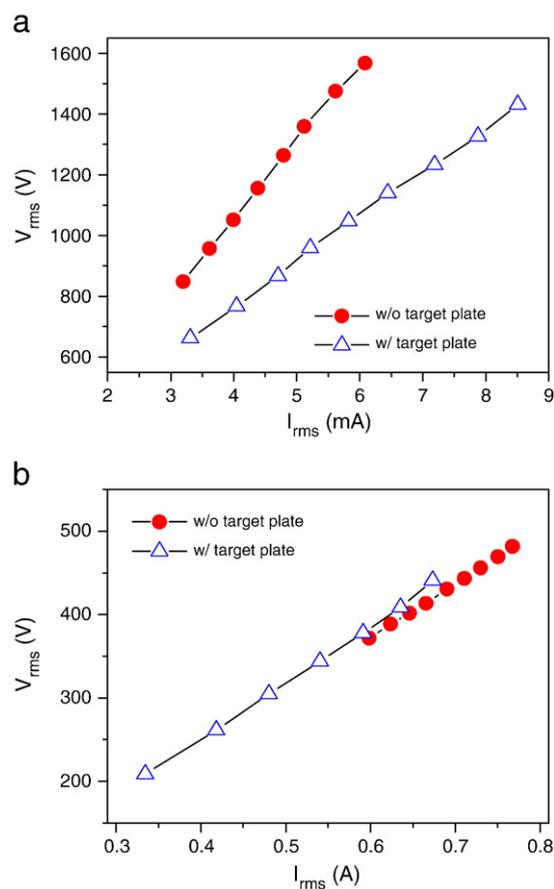


Fig. 2. I - V curves for (a) LF and (b) RF plasmas. \bullet - \bullet : without a target plate, \triangle - \triangle : with a target plate.

RF plasma conduction current with the plate was between 11 mA and 43 mA, while that without the plate was between 9 mA and 15 mA for the same discharge voltage range. Although the increase in the conduction current was much larger for the LF plasma, the RF conduction current is still about 4 times higher than the LF conduction current in the same dissipated power range. Yet, this lower density LF plasma has reactive oxygen species and is strong enough to inactivate microorganisms, as confirmed by our previous studies and by the work of the other research group [3,18].

3.2. Surface treatment of pork

Simulating the plasma surface treatment of biomaterials, pork samples were prepared and treated with the plasma. The pork samples had an average thickness of about 1 cm. The DC resistance of the pork samples varied between 0.5 M Ω and 3 M Ω , depending on the water contents. The pork sample consists of water (70%), protein (20%), fat (6%), carbohydrate, mineral, etc. (the amount varies depending on the part). Due to the lower resistance or the higher conductivity of the pork sample compared to the ITO plate, the plasma underwent arc transition at a threshold current. Hence, the pork plasma treatment was done where the plasma was in the glow mode.

The pork sample was placed on a grounded ITO plate. Fig. 3(a) and (b) depicts the I - V curve and the gas temperature during the pork treatment using the LF plasma, respectively. Similarly, Fig. 3(c) and (d) depicts the I - V curve and the gas temperature using the RF plasma. The conduction current through the pork sample was less than 0.5 mA and that of RF plasma was less than 40 mA. As given by the current values, the LF plasma conduction current is low enough not to cause electrical

damage to the living tissue; however, the RF plasma conduction current is around the electric shock safety limit [23]. Nevertheless, the RF plasma did not transit to arc, because the higher frequency prevents the transition to arc at lower current [24]. The LF plasma gas temperature was almost constant at 22 °C when the room temperature was 23 °C, and the RF plasma gas temperature was started from 26 °C and then increased to 31 °C. The LF plasma gas temperature was changed little from the single electrode plasma case, but the RF plasma gas temperature was about 2.5 °C higher for the same discharge.

The plate under the pork sample was changed from ITO to copper and then compared with the ITO case. For both LF and RF cases, the conducting copper allowed the breakdown voltage to be decreased, because there was no voltage drop as in the ITO case. As a result, the discharge voltage and current regime where the plasma was in the glow mode during the pork treatment was shifted to the left (lower current region), as presented in Fig. 4. Interestingly, the shift is much larger for the RF plasma, and the discharge current was decreased by half. This can be explained by the higher electrical conductivity of pork for the higher frequency [25].

Another possible way to control the plasma operating condition is to change the distance between the pin electrode and the pork sample. In the case of the LF plasma, the plasma length was about an order longer than the RF plasma. Thus, the LF plasma treatment could be done at a much greater distance from the pin electrode. The greater distance made the plasma pork treatment occur at a higher discharge voltage and current by reducing the electric field over the space between the pin and the pork. Also, as the glow to arc transition threshold current was raised, the glow mode operating range was increased.

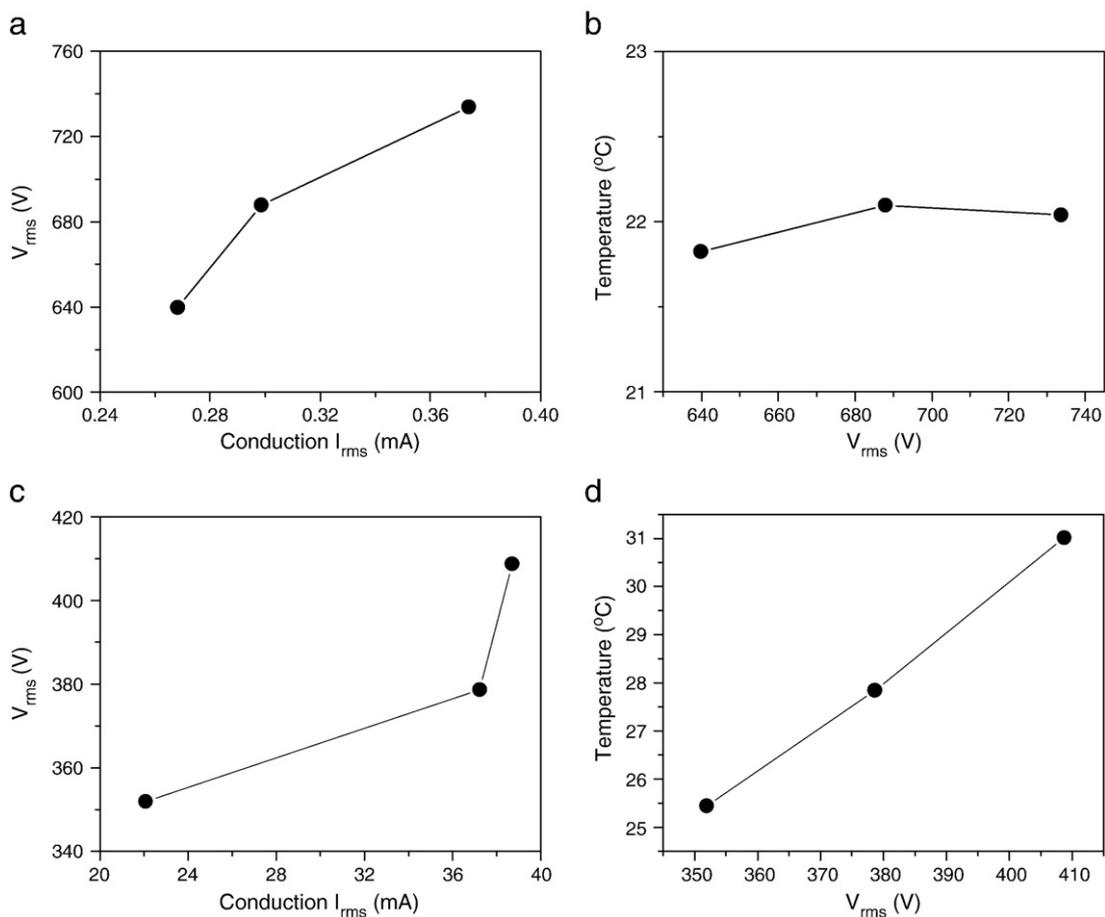


Fig. 3. I - V curves for (a) LF and (c) RF plasmas and plasma gas temperature plots for (b) LF and (d) RF plasmas during the pork surface treatments.

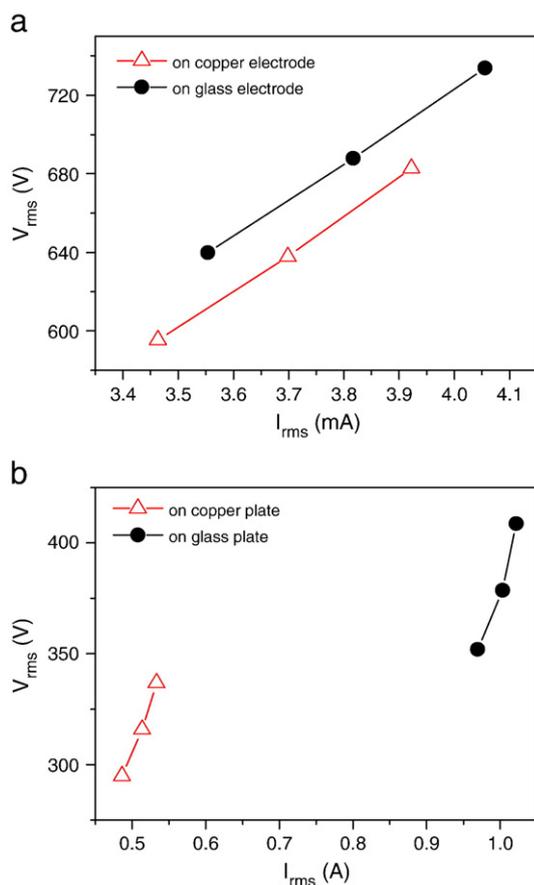


Fig. 4. I - V curves for (a) LF and (b) RF plasmas —●—: on a glass plate, —△—: on a copper plate.

4. Summary

A target plate made of ITO glass was added to a single pin electrode discharge system that employed either an LF (50 kHz) or RF (13.56 MHz) power source. The LF plasma was more significantly affected by its presence due to the limited dielectric effects in the RF range. As a result, the LF plasma conduction current was increased by almost fourteen times while the RF plasma conduction current was increased by less than three times.

The feasibility of plasma biomaterial surface treatments was studied using a pork sample. During treatment of the pork, the conduction current of the LF plasma was less than 0.5 mA, while that of the RF plasma was close to the electrical shock limit for humans, at less than 40 mA. The LF plasma gas temperature was almost constant at 22 °C, and the RF plasma gas temperature was started from 26 °C and then increased to 31 °C. The substrate material under the pork was changed to copper, and due to the higher conductivity of the pork at the higher frequency, the discharge regime was shifted far toward the lower current region. In addition, the much shorter jet size of the RF plasma can only treat the pork sample from distances of a few millimeters, while the LF plasma can treat the pork sample from a distance of several tens of millimeters. At such a long distance, the plasma was electrically safer as it did not transit to arc for a higher discharge current.

Acknowledgement

This work was supported by KAIST.

References

- [1] S.Y. Moon, W. Choe, B.K. Kang, Appl. Phys. Lett. 84 (2004) 188.
- [2] S.Y. Moon, W. Choe, H.S. Uhm, Y.S. Hwang, J.J. Choi, Phys. Plasmas 9 (2002) 4045.
- [3] D.B. Kim, J.K. Rhee, B. Gweon, S.Y. Moon, W. Choe, Appl. Phys. Lett. 91 (2007) 151502.
- [4] S.Y. Moon, J.K. Rhee, D.B. Kim, W. Choe, Phys. Plasmas 13 (2006) 033502.
- [5] S.Y. Moon, W. Choe, Spectrochim. Acta, Part B 58 (2003) 249.
- [6] S.Y. Moon, D.B. Kim, B. Gweon, W. Choe, Phys. Plasmas 15 (2008) 103504.
- [7] D.B. Kim, J.K. Rhee, S.Y. Moon, W. Choe, Thin Solid Films 515 (2007) 4913.
- [8] B. Gweon, D.B. Kim, S.Y. Moon, W. Choe, Curr. Appl. Phys. 9 (2009) 625.
- [9] E.E. Kunhardt, IEEE Trans. Plas. Sci. 28 (2000) 189.
- [10] K.H. Becker, K.H. Schoenbach, J.G. Eden, J. Phys. D 39 (2006) R55.
- [11] J. Franzke, K. Kunze, M. Miclea, K. Niemax, J. Anal. At. Spectrom. 18 (2003) 802.
- [12] K. Kelly-Wintenberg, A. Hodge, T.C. Montie, L. Deleanu, D. Sherman, J.R. Roth, P. Tsai, L. Wadsworth, J. Vac. Sci. Technol. A 17 (1999) 1539.
- [13] Y. Yang, Ind. Eng. Chem. Res. 41 (2002) 5918.
- [14] R.E.J. Sladek, E. Stoffels, J. Phys. D: Appl. Phys. 38 (2005) 1716.
- [15] E. Stoffels, I.E. Kieft, R.E.J. Sladek, J. Phys. D: Appl. Phys. 36 (2003) 2908.
- [16] A. Shashurin, M. Keidar, S. Bronnikov, R.A. Jurjus, M.A. Stepp, Appl. Phys. Lett. 93 (2008) 181501.
- [17] S. Yonson, S. Coulombe, V. L'èveillé, R.L. Leask, J. Phys. D: Appl. Phys. 39 (2006) 3508.
- [18] S.J. Kim, T.H. Chung, S.H. Bae, S.H. Leem, Appl. Phys. Lett. 94 (2009) 141502.
- [19] W. Meyer, Hautarzt 47 (1996) 178.
- [20] J.K. Lunney, Int. J. Biol. Sci. 3 (2007) 179.
- [21] U. Kogelschatz, Plasma Chem. Plasma Process. 23 (2003) 1.
- [22] S.Y. Moon, D.B. Kim, B. Gweon, W. Choe, Appl. Phys. Lett. 93 (2008) 221506.
- [23] C. Nave, B. Nave, Physics for the Health Sciences, Saunders, Saint Louis, Missouri, 1985.
- [24] J. Park, I. Henins, H.W. Herrmann, G.S. Selwyn, R.F. Hicks, J. Appl. Phys. 89 (2001) 20.
- [25] A. Sabouni, S. Noghianian, S. Pistorius, IFMBE Proceed. 3 (2006) 148.