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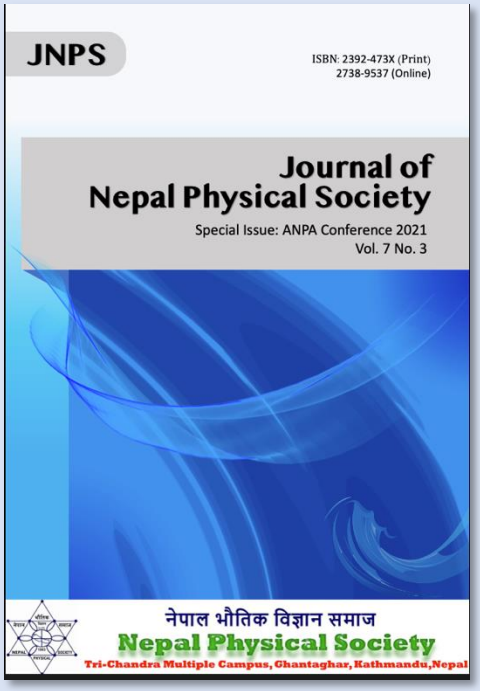

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GENERATION OF ATMOSPHERIC PRESSURE DIELECTRIC BARRIER DISCHARGE (DBD) USING WATER ELECTRODE

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Abstract. In this experiment, an atmospheric pressure dielectric barrier discharge (DBD) generated with a water electrode is investigated by means of optical measurements and imaging. The discharge was generated using a high voltage (0-20kV) power supply operating at 10-30 kHz with water as one of the electrode and borosilicate glass as a dielectric barrier of 2.5mm thickness. This paper reports the generation and characterization of atmospheric pressure plasma in nitrogen environment and its application in the surface modification of polyethylene terephthalate (PET). The generated plasma has been characterized by image analysis and optical emission spectroscopy. Our results showed that the distribution of micro-discharges depends significantly on the inter electrode gap and applied voltage. In order to characterize the discharge, electron temperature has been determined by using line intensity ratio method. The results showed that T_e depends on applied voltage and pressure inside the chamber. The values of T_e were found to be 1.40 eV and 0.95 eV applied at 1kVand 10kV voltage using 1% concentration. The discharge was produced at various conditions for the study of effectiveness of treatment on the surface property of Polyethylene terephthalate (PET). After the treatment of the sample in different treatment time: 10s, 20s, 40s, and 60s, the hydrophobic properties of sample changed to the hydrophilic. To investigate the effect of plasma treatment on Polyethylene terephthalate (PET) polymer contact angle was measured by using goniometer with water as a testing liquid. The surface properties of the untreated and plasma treated PET samples were characterized by contact angle measurement, and surface energy analysis. Before treatment the contact angle for untreated sample was 77.1° and after treatment its contact angle becomes 38.7°, 35.04°, 33.6° and 31.6° respectively.

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Keywords: Atmospheric pressure plasma; Electron temperature; Optical Emission Spectroscopy

INTRODUCTION

Nonthermal and low-temperature plasma have become a topic of interest to research. Development of various types of lower – pressure system and atmospheric – pressure system have already been developed. Polymer surface treatment, deposition of thin films and cleaning of substrates requires low-pressure plasma sources. [1,2].

Low – pressure plasma got more popularity due to its wider field of applications such as: polymer treatment, surface cleansing, pollution control, thin film deposition, biological sterilization, water purification and many more. It has also a great importance in manufacturing semi-conductor devices and in materials processing. [3,4,5] The surface treatment of polymers to enhance their hydrophilicity is one of the major areas of application of atmospheric pressure plasma. This implementation is motivated due to much superiority over the conventional method of surface treatment. [6, 7].

Polymers are generally macromolecules formed by the repeated linking of a large number of small

molecules [8]. Polymers which play a significant role as structural materials are generally utilized in industries [9, 10]. Due to their superior performance, low cost, good breakage resistance, transparency, and low inflammability polymers are fascinating business articles. Their surface modification is mandatory due to its low hardness, low scratch resistance, and degradation by UV radiation [11, 12]. With low surface energy, poor chemical reactivity, and a weak cohesion layer on the surface, it is vital to enhance the surface properties of polymers without changing the bulk properties [13, 14]. PET is an inert material and is widely accepted as a safe and recyclable plastic. Due to these reasons, it is widely used in beverage packaging, electronics, and biomedical industries. Similar to glass, it is hygienic and generally resistant to attack by bacteria and other microorganisms [15].

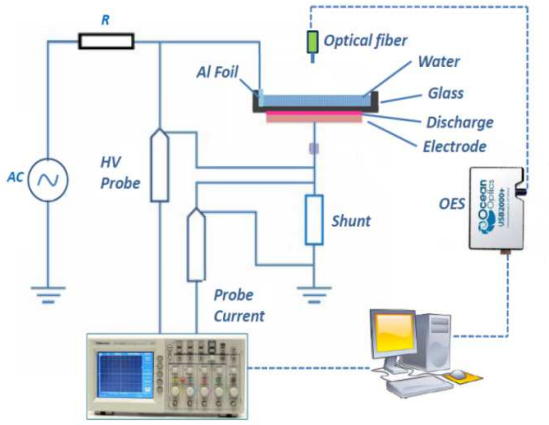


Figure 1: Schematic diagram of the experimental setup

DBD plasma with a liquid electrode in atmospheric air is scarce in scientific literature and a topic of an ongoing investigation. This is mainly due to plasmas in liquids are more tangled than their counterparts in the gas phase. They are in a highly non-equilibrium state and are mostly generated in both gas and liquid state. In particular species and charge transfer mechanisms at the plasma liquid interface and the intensive evaporation make these discharges a lot more complex. Discharges in and in contact with liquids produce intense UV radiation, shock waves, and several reactive chemical species (OH, atomic oxygen, hydrogen peroxide, etc) As a result plasma is in or in contact with liquids suitable for a large variety of potential applications. It becomes very effective in the treatment of many biological and chemical matters. This makes liquid plasmas particularly suitable for water remediation and polymer surface activation. That's why this kind of plasma has been studied extensively in the last two decades. Liquids were already introduced in the plasma field in the 20th century, so the field is not as new as often thought [16].

METHODS

The reactor was designed and fabricated at the Kathmandu University Plasma Physics laboratory. The experimental setup consists of a DBD system with water as one of the electrode and borosilicate glass as a dielectric barrier of 2.5mm thickness. The electrodes are connected to a high-voltage (0- 20kV) operating at (10-30) kHz frequency. The distance between two electrodes is fixed. Plasma was produced at atmospheric pressure in air. The schematic diagram of the experimental setup and the nature of the discharge are shown in Figure 1 and Figure 2.

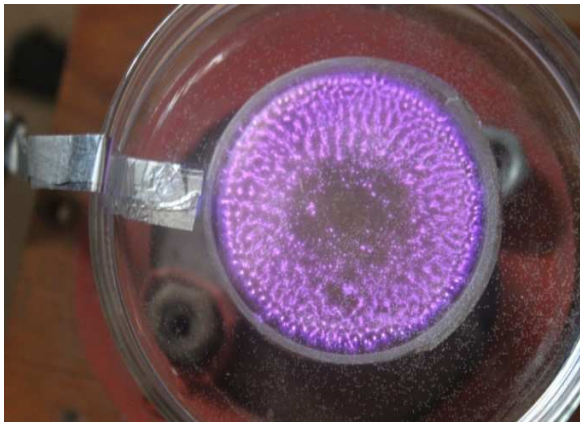


Figure 2 : Image of Discharge

Frequency is maintained at 27 kHz. . The discharge thus obtained was investigated by image analysis and optical emission spectroscopy. The optical characterization of the discharge has been done using the line intensity ratio method with the help of an optical emission spectrometer (USB 2000+, Ocean Optics). The contact angle of the untreated and plasma treated PET samples is measured using the Rame Hart contact angle goniometer (Model 200). Before the treatment, the samples of PET of dimension (50 mm × 15mm ×0.05mm) were taken.

The samples were provided by Good fellow, U.K. Before treatment, polymer samples were washed in

isopropyl alcohol in order to remove organic contaminants from the surface of the specimens. The samples were then ultrasonically cleaned in distilled water for 10 minutes and dried at room temperature. The contact angle measurement was done at five different locations of the same samples and the average value of the contact angle thus obtained was used for the surface energy calculations [17].

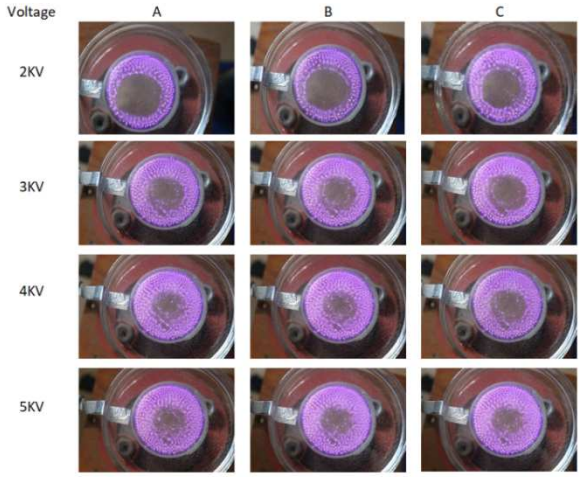


Figure 3: Three images of discharge A, B and C at 27 kHz by applying voltage of 2 kV, 3 kV, 4 kV and 5 kV respectively.

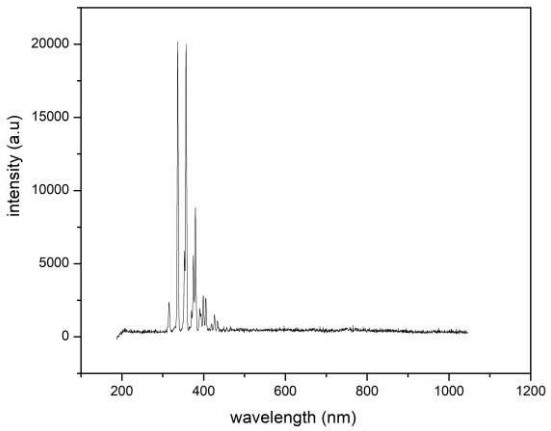
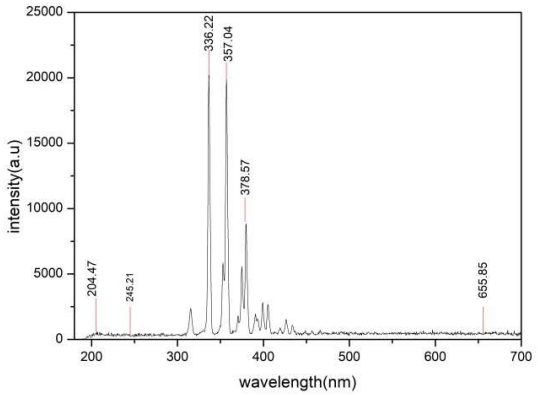
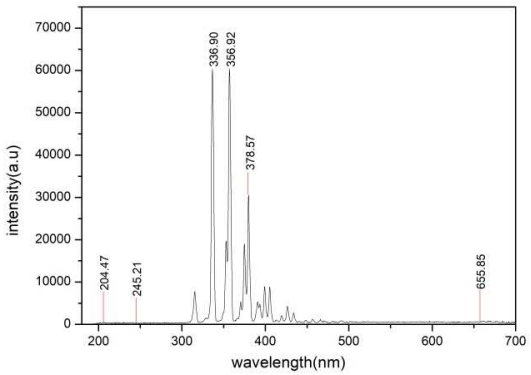


Figure 4: Optical Emission Spectroscopy of Atmospheric Pressure Dielectric Barrier Discharge.



(a)



(b)

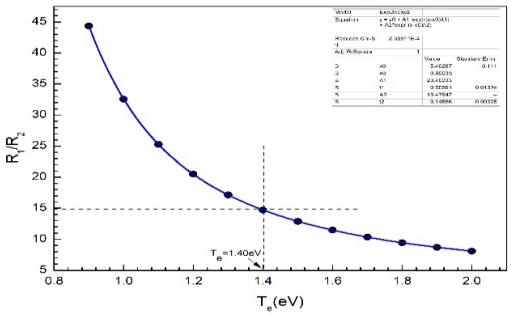
Figure 5: Spectra of APDBD in the range of 180nm to 700nm a) at 1kV having 1% HNO_3 solution; (b) at 10kV having 1% HNO_3 solution.

RESULTS AND DISCUSSION

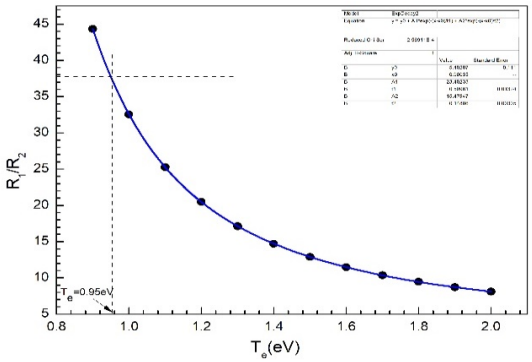
I. Image analysis: The discharge thus obtained was investigated by image analysis by using high resolution digital camera. Our results showed that the distribution of micro - discharges depends significantly on the inter electrode gap and applied voltage. Images of discharge at 27kHz using water electrode is shown in figure 3.

II. Optical Characterization of the Discharge:

Figure 4 shows the spectra of the discharge and their corresponding intensities and wavelength using



(a)



(b)

Figure 6: Plot of R_1/R_2 as a function of T_e applied voltage a) 1kV and b) 10kV having 1% HNO_3 solution.

atmospheric air. Figure 5 shows the spectra of APDBD in the range of 180nm to 700nm at 1kV and 10kV applied voltage using 1% HNO_3 solution. The optical characterization of the discharge was carried out by using the line intensity ratio method. In this method, four suitable lines (two for NII and two for NIII) are chosen and electron temperatures is estimated using the lines intensity ratio method obtained from the discharge. The working formula used to calculate the electron temperature is as follows

$$\frac{R_1}{R_2} = \frac{\frac{I_1}{I_2}}{\frac{I_3}{I_4}} = \left(\frac{A_{pq}}{A_{rs}}\right)\left(\frac{g_p}{g_r}\right)\left(\frac{\lambda_{rs}}{\lambda_{pq}}\right)\left(\frac{A_{uv}}{A_{xy}}\right) \qquad (1)$$

$$\left(\frac{g_u}{g_x}\right)\left(\frac{\lambda_{xy}}{\lambda_{uv}}\right) Exp\left[-\frac{E_p - E_r - E_x + E_u}{KT_e}\right]$$

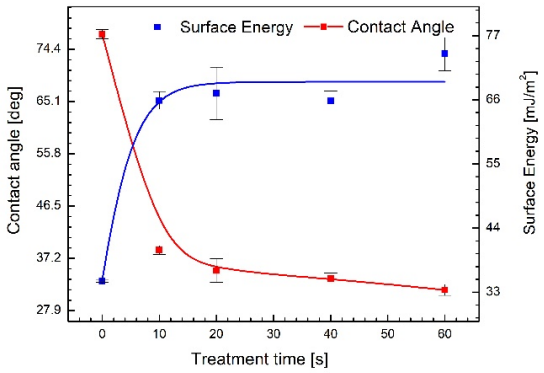


Figure 7: Contact angle and surface energy as a function of the treatment time in the discharge.

Here, in Equation (1), R is the ratio of the intensity of two lines, I is the intensity of the spectral line, A_{ij} is the transition probability of the transition i → j, g_i is the statistical weight of the upper level, λ is the wavelength of the line radiation, E_i is the energy of the upper level, KB is the Boltzmann constant, and T_e is the electron temperature. The values of λ and I are obtained from the observation, and the values of A_{ji}, g_i, and E_i are obtained from the National Institute of Standards and Technology (NIST) Atomic Spectra Database.The corresponding values of the transition probability, statistical weight, and energy levels for the Nitrogen II and Nitrogen III lines were obtained through the NIST database [18]. Table 1 shows the corresponding value of the ratio of the intensity of the spectral lines (R_1/R_2) with change in electron temperature (T_e).

Table 1: Values of R_1/R_2 for different T_e

Electron temperature T_e(eV)	Intensity ratio R_1/R_2
0.9	44.34
1.0	32.56
1..1	25.28
1.2	20.48
1.3	17.14
1.4	14.71
1.5	12.88
1.6	11.47
1.7	10.36
1.8	9.46
1.9	8.72
2.0	8.10

From Figure 6, the electron temperature (T_e) was found to be 1.40 eV for applied voltage 1kV and0.95 eV for applied voltage 10 kV using 1% HNO_3 solution respectively.

III. Surface Modification of Polyethylene Terephthalate (PET)

Contact Angle and Surface Energy Measurements

For an ideal, smooth, and homogeneous surface, the water contact angle and surface free energy are measured at the equilibrium according to Young's equation and Fowke's equation respectively [19] [20].

$$Cos\theta = \frac{\gamma_{sv} - \gamma_{sl}}{\gamma_{lv}} \qquad (2)$$

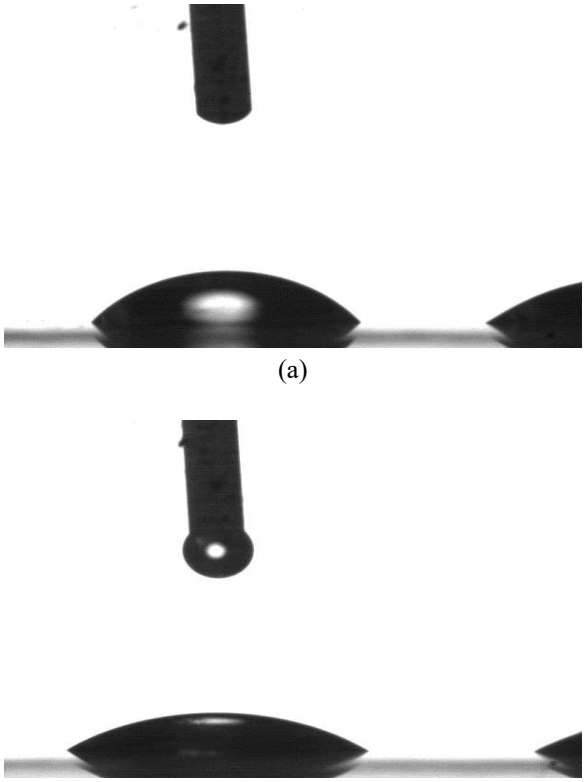
where γ_{sv} is the surface free energy of the solid substrate, γ_{sl} is the interfacial tension between the solid and the liquid, and γ_{lv} is the surface tension of the liquid. For two liquids *i* and *j*,

$$\gamma_{li}(1 + Cos\theta_i) = 2[\gamma_{li}^d \gamma_s^d]^{\frac{1}{2}} + 2[\gamma_{li}^p \gamma_s^p]^{1/2} \qquad (3)$$

$$\gamma_{lj}(1 + Cos\theta_j) = 2[\gamma_{lj}^d \gamma_s^d]^{\frac{1}{2}} + 2[\gamma_{lj}^p \gamma_s^p]^{1/2} \qquad (4)$$

Substituting the known values of the surface tension and its polar and dispersion components of the test liquids, components of surface free energy of the solid γ_s^p and γ_s^d can be determined by solving Equations (3) and (4). The sum of these two quantities eventually gives the total surface energy of the solid.

The treatment of the sample was performed for various exposure times (0 - 60 seconds). The effect of this parameter on hydrophilicity was scrutinized by contact angle measurement with water as a testing liquid on the surface. It is seen that a rapid decrease in the static water contact angle takes place with the treatment time up to 20 seconds which shows an increase in wettability in the surface induced by APDBD treatment. Initially, the contact angle of the untreated PET for water was 77.1°, but after plasma treatment, the contact angle was effectively reduced to 35.04° and became almost constant after 20 seconds of treatment time as shown in Figure 7. Images of water drops on PET polymer (untreated and treated) are shown in figure 8. The reduction in contact angle might be due to the change in the surface roughness. [21]



(a)

(b)

Figure 8: Images of water drops on PET polymer (a) Untreated (b) Treated

The variation of surface free energy and its various polar and dispersive components with treatment time for PET polymer is shown in Figure 9.

As seen in Figure 9, the surface energy of the untreated PET was found to be 34.9 mJ/m^2. It is seen that the surface energy significantly increases for a few second of treatment time, becomes maximum to about 73.9 mJ/m^2 after 20 seconds. Similar trend is also observed for the polar component and it is mainly due to the incorporation of the polar species such as carbonyl (C=O), hydroxyl (-OH) and carboxyl (-COOH) groups on the treated PET surface. The dispersive component decreases initially and remains almost constant which attributes to more physical change than chemical change.

This component does not have any contribution to increase the wettability of the PET surface [17].

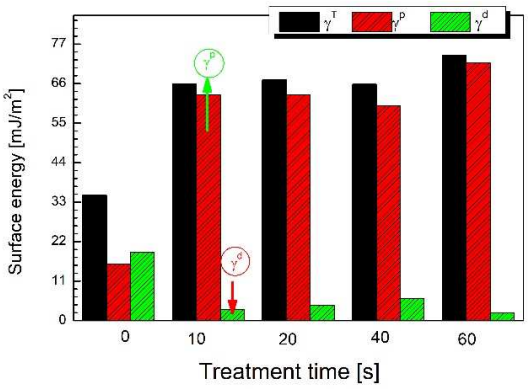


Figure 9: Variation of surface free energy with treatment time for PET samples.

CONCLUSIONS

The atmospheric-pressure plasma has been produced and characterized by image analysis and optical methods. DBD with a water electrode system facilitates the direct observation of the front view of the discharge which makes it possible to investigate the uniformity of the micro-discharges over the electrode surface. Our results showed that the distribution of micro - discharges depends significantly on the inter electrode gap and applied voltage. Here the number of micro discharges increases with the increase in applied voltage.

Electron temperature (T_e) were found to be 1.40 eV and 0.95 eV applied at 1kVand 10kV voltage using 1% concentration, respectively, using the line intensity ratio method. In general, electron temperature T_e increases with increase applied voltage. But in this study, we observe that the electron temperature decreases slightly with an increase in the applied voltage. There is no any significant change in this experiment. Although this result is unlikely, it has good agreement with the results reported by Validity of electron Temperature, Measurement by Using Boltzmann Plot Method in Radio Frequency Inductive Discharge in the Atmospheric Pressure Range.

Treatment of PET using the atmospheric-pressure plasma resulted in an improvement in hydrophilicity. It is mainly due to the increase in the polar component of the surface free energy after plasma treatment which indicates the formation of polar functional groups on the surface. The improvement of the wettability of PET strongly depends on the treatment time. Results showed that there is a significant reduction in the water contact angle on the polymer surface after plasma treatment with a consequent increase in its surface free energy.

AUTHOR CONTRIBUTIONS

Deepak Prasad Subedi presented the idea for the paper. Roshna Manandhar carried out the methodology, data processing, experimental methodology and draft preparation; Rajesh Prakash Guragain helped in the analytical methods and experimental set up. Rabin Baral provided computational resources and carried out manuscript editing tasks. Finally, Deepak Prasad Subedi contributed to the final version of the manuscript.

EDITOR'S NOTE

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