Surface Treatment of Handmade Lokta Paper by 50 Hz Dielectric Barrier Discharge Generated in Air at Atmospheric Pressure for Improvement of Hydrophilicity

G. K. Chhetri[1*], R. P. Guragain[1*], U. M. Joshi[1], and D. P. Subedi[1]

[1] *Department of Physics, School of Science, Kathmandu University, Dhulikhel, Nepal*

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The purpose of the current study is to characterize 50 Hz dielectric barrier discharge generated in the air at atmospheric pressure using a sinusoidal voltage of 12.8 kV (rms) by performing electrical and optical diagnostics and to investigate its effects on the improvement of hydrophilicity and weight loss (%) of handmade Lokta paper. The energy dissipated per cycle, and the average power dissipated was determined from Lissajous figures. The estimated value of electron density and electron temperature in the discharge were 8.47×10^{8} cm^{-3} and 1.29 eV. 50 Hz air DBD can be used for long-term operations and continuous treatment of Lokta paper. The effects of the discharge on the hydrophilicity and weight loss (%) of Lokta paper were studied respectively by the wicking test and gravimetric method. The results showed a significant improvement in the hydrophilicity and considerable weight loss (%) in Lokta paper after the plasma treatment. The increase in hydrophilicity of Lokta paper gives an enormous possibility for the quality and colorful designs of the Handmade Lokta paper and widens the area of its uses.

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*ganes.kc7@gmail.com, *rayessprakash@gmail.com

I. Introduction

The real start of the knowledge era in human history is regarded to have been possible only after the invention of paper. People had reflected their talent and imaginative power by drawing different images on the cave walls, rocks, and dry leaves even before the paper was invented. The invention of paper made it possible to write books, keep printed records of various documents and even print the most convenient form of money. The invention of paper is believed to have taken place first in China around 2000 years before.

Handmade Lokta paper was first produced in Nepal in the 12th century AD [1] and it has continuously been produced in Nepal since then. The plant, which grows as a wild shrub in the high Himalayan forests with an altitude of "1,600 m to 4,000 m" [2,3], is locally known as Lokta (Daphne papyracea and Daphne bholua). This plant is the source of Lokta paper (Nepali Kagaj). The Lokta plant is not cultivated as it occurs naturally in the forests.

In modern times, almost all the printers and publishers have been using machine-made papers; however, the use of handmade Lokta paper is still wide in the form of art paper, attractive stationery, and handicrafts. Moreover, the handmade Lokta paper is commonly used for producing birth certificates, land ownership papers, and all forms of judicial documents. Lokta paper equally carries the religious value as it was used in the Buddhist monasteries to write, print, and draw the teachings of Lord Buddha. In addition to these, there are various other fashionable products made from Lokta paper such as diaries, notebooks, lampshades, writing sets, photo albums and frames, gift boxes, greeting cards, wrapping papers, wallpaper, and many more decorative items.

Lokta paper is known for its durability and natural resistance to insects. The Lokta fiber is possibly longest and strongest fibers in Nepal. Handmade Lokta paper, therefore, is a very strong paper to make long-lasting paper products. This paper gets a distinct texture because of the uneven distribution and length of Lokta fibers. The processes of manufacturing Lokta paper such as cutting technique, pasting, and binding have been improving every new day. The production of a larger variety of color papers has increased due to the modern technique known as ‘dip dying’. In the same way, the calendaring process has assisted to prepare a smoother surface of Lokta paper for writing and printing work [4]. Besides, the production of the best quality of handmade Lokta paper requires some more technological innovations.

Several researchers have been working on the plasma treatment of paper. Plasma applications have been made for many years in the paper and packaging industries. Different plasma techniques have been tested to alter the surface energy of paper, functionalize surfaces, and improve their adhesion to various polymeric substrates. Deslandes et al. [5] used low-pressure radio-frequency nitrogen plasma to modify the surface of pure cellulose paper. They found measurable chemical changes in the surface composition and increased hydrophobicity of Whatman filter paper even after the very brief plasma exposure. Vohrer et al. [6] treated naturally aged ground wood paper using hydrogen after-glow plasma. They found a positive effect on the paper stability and measured reduction in microorganisms after the plasma treatment. Laguardia [7] focused their research work on the influences of plasma treatments on deteriorated ancient papers. They achieved a reduction of microbial contamination using plasma treatments. They showed that the use of the O_2/H_2 mixture increases the paper stability. Pykönen [8] studied the influence of plasma modification on surface properties and offset printability of coated paper. The author used DBD-based laboratory-scale and pilot-scale plasma activation equipment to modify the coated paper surface. He found plasma surface modification as an effective way to influence the paper surface energy and chemical composition. His results showed that plasma activation increased the surface energy and hydrophilicity of paper but decayed with time. He also found that hydrocarbon-, organosilicon- and fluorocarbon-plasma coatings provide hydrophobic character to the pigment-coated paper and the hydrophobic coatings were relatively stable with time. Galmiz et al. [9] studied the effects of surface dielectric barrier discharge on the surface properties of clay-coated paper. Their results showed considerable changes in wettability without significant modification of the morphology and surface structure of the paper after the plasma treatment in nitrogen as well as in ambient air.

In the present work, dielectric barrier discharge generated in the air at atmospheric pressure using 50 Hz line frequency is used for surface treatment of Lokta paper. It provides a convenient, cost-effective, and reliable technology to create non equilibrium plasma conditions in atmospheric pressure gases [10]. It has been used in several industrial applications for surface modification, including the textile, paper and packaging industries [11]. Plasma treatment affects the surface physically and chemically without changing its bulk properties [12–14]. The DBD reactor used here does not require any vacuum pump as in low-pressure plasma. That is why it can be easily scaled up for continuous treatment of paper for long-term operations and large-scale production. This study is mainly focused on wicking properties and weight loss (%) of Lokta paper. In 50 Hz DBD plasma treatment, the effect of treatment time on Lokta paper has been investigated. The wicking has been characterized by a vertical wicking test. Improvement on hydrophilicity and considerable weight loss (%) in Lokta paper gives an enormous possibility for the quality and colorful designs of the Lokta paper as well as its uses in other areas such as for paper towels, facial tissues, hydrophilic membrane, and hydrophilic filters in the liquid filtration process.

II. Experimental

II.a) Handmade Lokta Paper

In this study, the Lokta paper with thickness 0.204 mm and surface density 7.49 mg/cm^2 (Fig. 1) was obtained from the local market of Banepa, Baghmati Province, Nepal. All the samples were placed at ambient environment conditions before use.

Figure 1: Photograph of Handmade Lokta Paper

II.b) Atmospheric Pressure DBD Plasma Treatment

The Lokta paper was treated with atmospheric pressure DBD air plasma. Fig. 2 depicts a schematic figure of the experimental arrangement.

Fig. 3 shows a photograph of the DBD reactor. The DBD electrode configuration was positioned inside the transparent polycarbonate chamber (35.7 cm × 20.0 cm × 15.0 cm). The AC high voltage of 12.8 kV and frequency 50 HZ was applied across electrodes. The separation between the upper electrode (7.53 cm × 4.97 cm × 0.47 cm) and the grounded electrode (7.54 cm × 4.99 cm × 0.48 cm) is 0.35 cm. The dielectric barrier is polycarbonate plate (13.0 cm × 10.0 cm × 0.197 cm). The discharge was generated between two rectangular parallel electrodes. The experiment was carried out at ambient conditions.

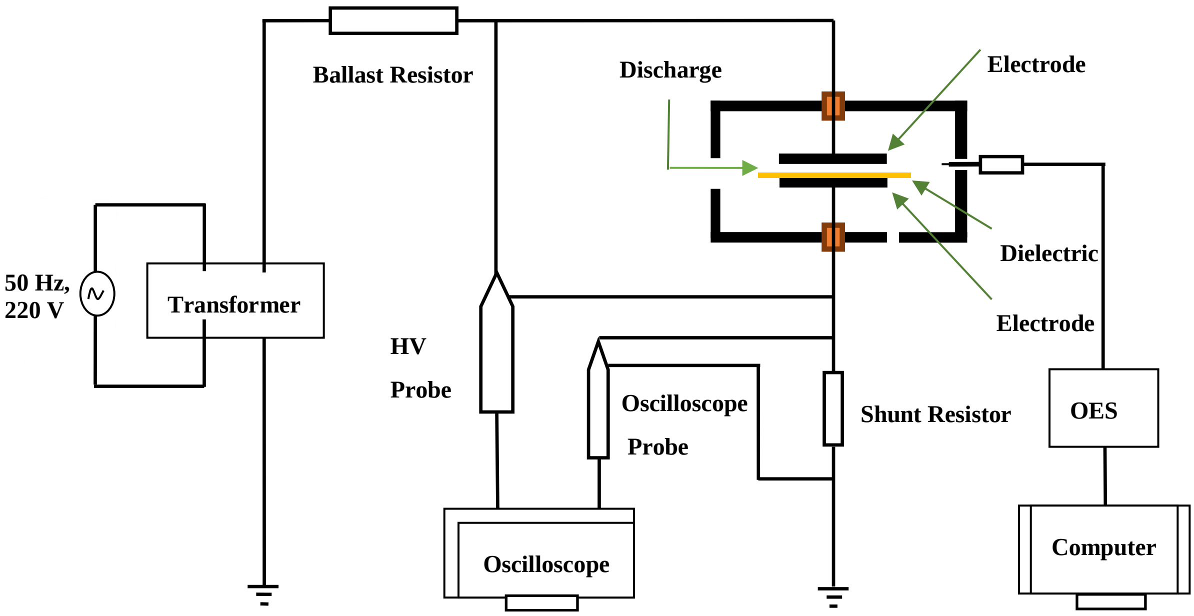

Figure 2: Schematic diagram of the experimental arrangement

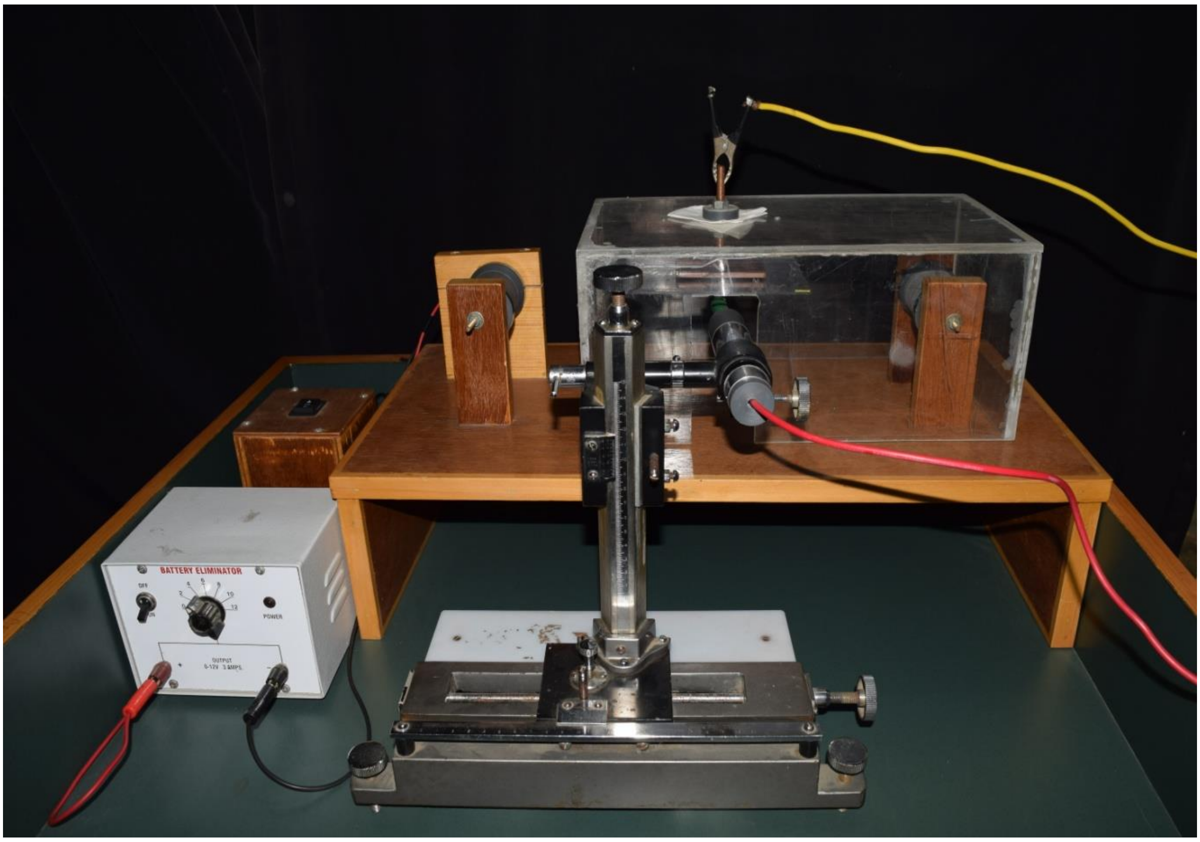

Figure 3: Photograph of DBD reactor

Fig. 4 shows the image of the discharge. A digital oscilloscope (TDS 2002, Tektronix), a high voltage probe (PINTEK HVP-28HF), and an oscilloscope probe are used to record the applied voltage and discharge current. An optical emission spectrometer [Ocean optics (USB+2000)] were used to record the emission spectra. A Nikon Digital Camera D3300 was used to get the image of the discharge.

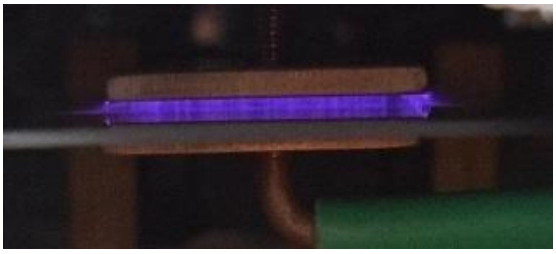

Figure 4: Image of the discharge

II.c) Wicking Test

The paper specimens of size 10 cm × 1.5 cm were prepared from Lokta paper. A 2B pencil was used to mark the scales of 9 cm in the specimen. The specimen was held vertically, and the lower edge was immersed in triple Deionized water (specific conductivity < 1) with 1cm of the sample below the water. The time when the water reached each graduated scale through the phenomenon of capillarity was recorded. The wicking test was performed under ambient environment conditions (Humidity: 51%; Temperature: 26^{0}C; Dew Point: 14^{0}C; Pressure: 10009 m Bar; UV Index: Very High, 9; Visibility 13 km) within 10 minutes after the plasma treatment. Four wicking test measurements were performed per sample per plasma treatment.

Fig. 5 depicts the experimental system for the measurement of vertical wicking.

Capillary penetration in the paper voids is the most important wetting mechanism [15]. This phenomenon can be illustrated by the Lucas-Washburn equation [16]:

$$l = \sqrt{\frac{2r\gamma\cos\theta + P_E r^2}{4\eta}}\ \sqrt{t} \qquad (1)$$

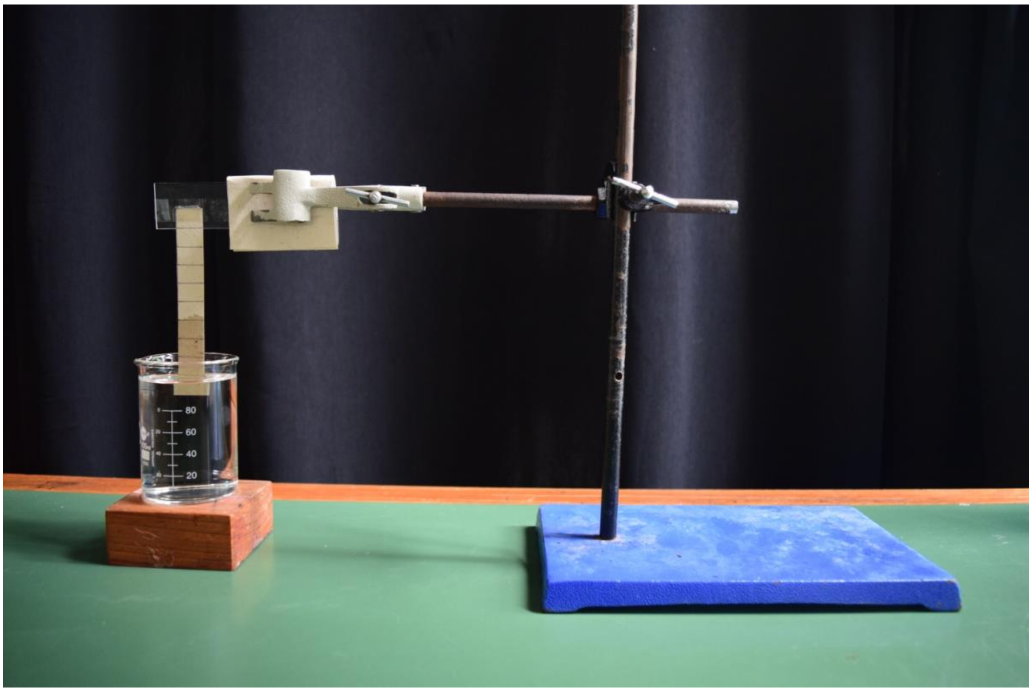

Figure 5: Water wicking in a typical vertical wicking test

where l is the length of penetration, P_E is the external pressure on the paper, r is the pore size of the paper, θ is the water contact angle of the paper, γ is the surface tension of the water, η is the viscosity of the water, and t is the time of penetration.

Equation (1) can be written as:

$$t_{\text{sqrt}} = \left(W_C^R\right) l \qquad (2)$$

where $t_{\text{sqrt}} = \sqrt{t}$, $W_C^R = \dfrac{1}{W_C}$, and $W_c = \sqrt{\dfrac{2r\gamma\cos\theta + P_E r^2}{4\eta}}$ is the wicking coefficient. The higher the W_c, the better is the water absorption ability [17].

II.d) Weight Loss (%)

The paper specimen of size 7.0 cm × 5.0 cm is prepared. An electronic precision balance (Bell Engineering, Model: MG124Ai) was used to measure the weight of paper specimens before and after the plasma treatment. The weight loss (%) of the plasma-treated paper was calculated by equation [18]

$$\text{Weight Loss (\%)} = \frac{W_1 - W_2}{W_1} \times 100\ \% \qquad (3)$$

where W_1 and W_2 are the weights of the paper before and after plasma treatment respectively. The weight loss is due to the prompt deduction of contamination from the upper surface of the paper. This is because of the energetic interaction of ions on the upper surface of the paper. The weight loss (%) in the treated paper is calculated by performing four experimental measurements per sample per plasma treatment time.

III. Results and Discussion:

III.a) Electrical Characterization:

Fig. 6 shows the typically applied voltage and discharge current waveforms in parallel-plate DBD. The electrical breakdown of the air gap in a DBD begins almost at the same time at numerous points of the surface and keeps on through the development of micro discharges. The existence of micro-discharge is a characteristics aspect of parallel plate DBD produced in the air at atmospheric pressure [19].

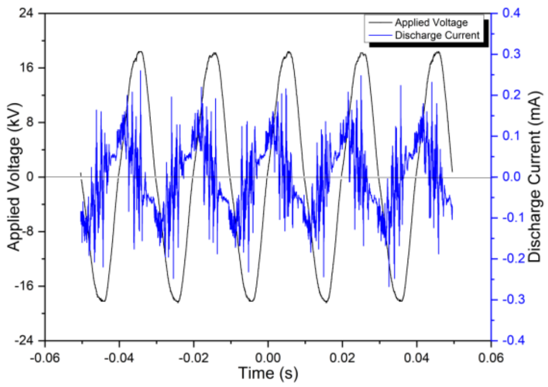

Figure 6: Typical current and voltage waveforms for the discharge

Fig. 7 depicts a typical charge (Q) – applied voltage (V) plot, in the course of one period of applied voltage, known as the Lissajous figure. The capacitance of the solid dielectric barrier, the equivalent capacitances between the electrodes, the energy deposited into the discharge during one cycle of applied voltage, and the average power deposited can be calculated employing Lissajous figures [20].

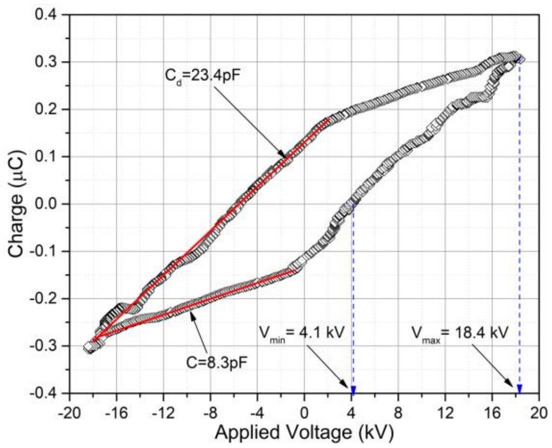

Figure 7: A characteristic Lissajous figure for the DBD in the air with applied voltage (V) = 12.8 kV (rms) and frequency (f) = 50 Hz.

The equivalent capacitance between the parallel electrodes for volume discharge, C is calculated by [21]:

$$\frac{1}{C}=\frac{1}{C_{air}}+\frac{1}{C_{die}} \quad (4)$$

Where, C_{air} is the capacitance of the air gap and C_{die} is the capacitance of dielectric.The capacitance of the dielectric (*C_{die}*) is 23.4 pF. The equivalent capacitance between the electrodes (*C*) is 8.3 pF. The capacitance of the air gap (*C_{air}*) is 12.9 pF.

The energy deposited into the discharge through one cycle of the applied voltage *E* is calculated by [22]:

$$E=4C_{die}\frac{1}{1+\frac{C_{air}}{C_{die}}}\mathrm{V}_{\min}(\mathrm{V}_{\max}-\mathrm{V}_{\min}) \quad (5)$$

and mean power is calculated [23] :

$$P=4fC_{die}\frac{1}{1+\frac{C_{air}}{C_{die}}}\mathrm{V}_{\min}(\mathrm{V}_{\max}-\mathrm{V}_{\min}) \quad (6)$$

where V_{min} is the lowest value of applied voltage necessary to ignite the discharge, Vmax is the highest value of applied voltage and f is the frequency of applied voltage. The energy deposited into the discharge through one cycle of applied voltage is 3.54 mJ and the mean power is 177 mW. The electron density (n_e) can be calculated by an equation [24]:

$$\mathrm{J}^{e}_{\mathrm{cond}}=-n_e e\mu_e \mathrm{E} \quad (7)$$

Where $\mathrm{J}^{e}_{\mathrm{av}}$ is the average current density, e is the electronic charge, μ_e is the electron mobility and E is the electric field in the discharge region.In this work, the average current density is 1.08×10^{-3} A/cm^2, the electric field strength across the discharge gap (E) is 1.72 x 10^4 V/cm, electron mobility (μ_e) estimated from BOLSIG + software is 462 cm^2/Vs and the electronic charge (e) is 1.6 x 10^{-19} C. Using all these values in Eq. (7), the electron density (n_e) is found to be 8.47×10^8cm^{-3}.

III.b) Optical Characterization

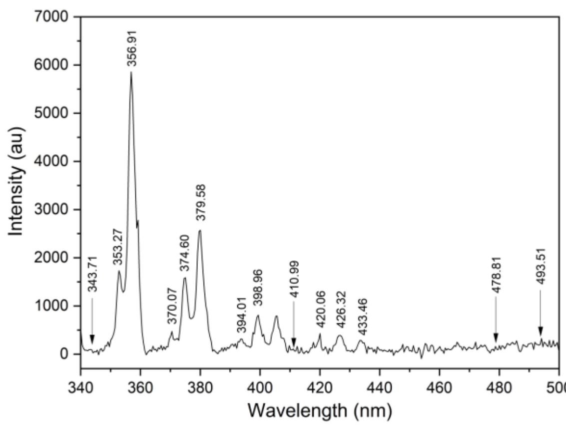

Figure 8: Optical emission spectra of APDBD in air

Fig. 8 shows the optical emission spectra (OES) from the 50 Hz atmospheric pressure DBD generated in air at atmospheric pressure.

The line intensity ratio method was used for the measurement of the electron temperature [25,26]:

$$\frac{R_1}{R_2}=\frac{I_1/I_2}{I_3/I_4}=\left(\frac{A_{pq}}{A_{rs}}\right)\left(\frac{g_p}{g_r}\right)\left(\frac{\lambda_{rs}}{\lambda_{pq}}\right)\left(\frac{A_{uv}}{A_{xy}}\right)\left(\frac{g_u}{g_x}\right)\left(\frac{\lambda_{xy}}{\lambda_{uv}}\right)\exp\left[-\frac{E_p-E_r-E_x+E_u}{kT_e}\right] \tag{8}$$

where 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, k is the Boltzmann constant and T_e is the electron temperature.

Two NI lines (493.51 nm, 410.99nm) and two N II lines (478.81nm, 343.71nm) were chosen from the discharge spectra. The corresponding values of A_{ji}, g_i, λ and E_i were taken from the National Institute of Standards and Technology (NIST) Atomic Spectra Database Lines Data [27], and the values of λ and I are obtained from the discharge spectra to measure the electron temperature T_e. Tab. 1 presents the ratios of spectral lines (R_1/R_2) for different electron temperatures T_e.

Electron Temperature (T_e)	Ratio of Spectral lines (R_1/R_2)
0.8	2.08
0.9	1.65
1.0	1.38
1.1	1.19
1.2	1.05
1.3	0.94
1.4	0.86
1.5	0.79
1.6	0.74
1.7	0.70
1.8	0.66
1.9	0.63
2.0	0.61

Table 1: Ratios of spectral lines for different electron temperatures.

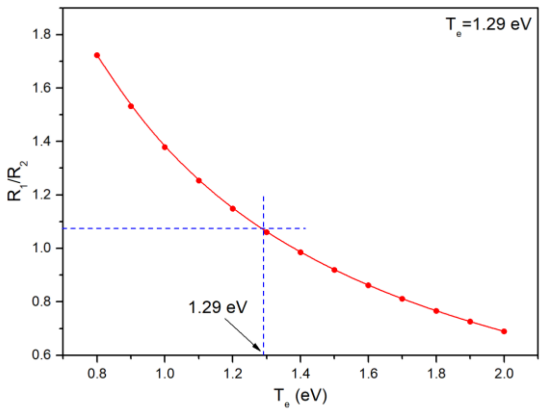

Figure 9: Plot of R_1/R_2 as a function of T_e

From Fig. 9, the electron temperature (T_e) is estimated to be 1.29 eV.

3.3 Wicking Performance of Lokta Paper

Fig. 10 shows the wicking rate of untreated, and plasma treated Lokta paper. The wicking time versus wicking height for different treatment time is plotted to illustrate the comparison [28–30]. It is observed that the wicking time to reach 7 cm height for untreated paper is 786 s and it is reduced to 486 s when the plasma treatment is carried for about 2 minutes. It is observed from the graph that all plasma treated paper has considerable enhancement in wicking rate when compared with untreated paper. There is not much change in the wicking time between 2 and 10-minutes plasma-treated samples.

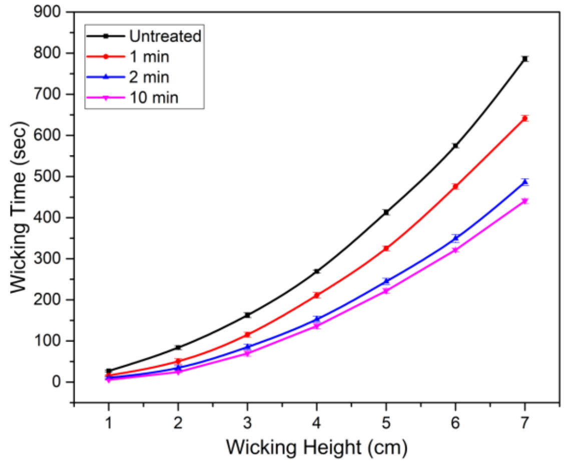

Figure 10: Wicking time of untreated and plasma treated Lokta paper at constant voltage 12.8 kV (rms) with different treatment time

Fig. 11 shows the square root of time of penetration *(t_{sqrt})* versus length of penetration (*l*) plot for different exposure time. The wicking coefficient (W_C) for untreated sample was 0.246. The wicking coefficient (W_C) for 1 minute, 2 minutes and 10 minutes plasma-treated samples were respectively 0.276, 0.316, and 0.339. The wicking coefficients of 2 minutes and 10 minutes plasma treated sample were respectively 1.28 times and 1.37 times greater than the wicking coefficient of the untreated sample. It is found that all plasma-treated paper has a significant increment in water absorption ability compared with the untreated sample. The results have indicated that 2 minutes of treatment time is sufficient to increase the absorption ability of paper significantly.

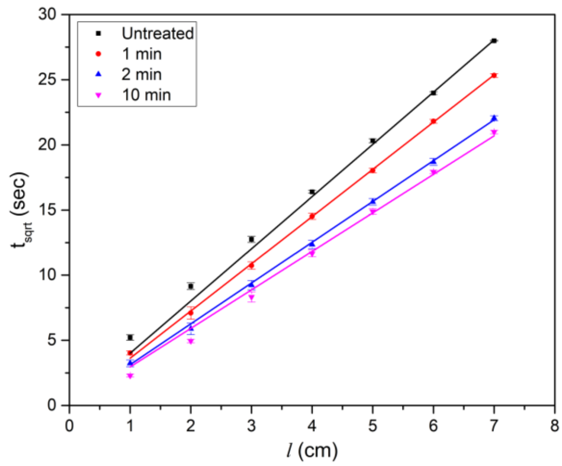

Figure 11: Wicking coefficient of untreated and plasma treated Lokta paper at constant voltage 12.8 kV (rms) at different exposure time

III.c) Weight Loss (%):

Fig. 12 shows the weight loss (%) of Lokta paper after the plasma treatment at different treatment time. It has been observed that the weight loss (%) is slightly increased with the treatment time from 1 minute to 3 minutes, and then remains nearly constant. The weight loss might be due to the cleaning effect and etching effect of cold plasma on the upper surface layer of the paper.

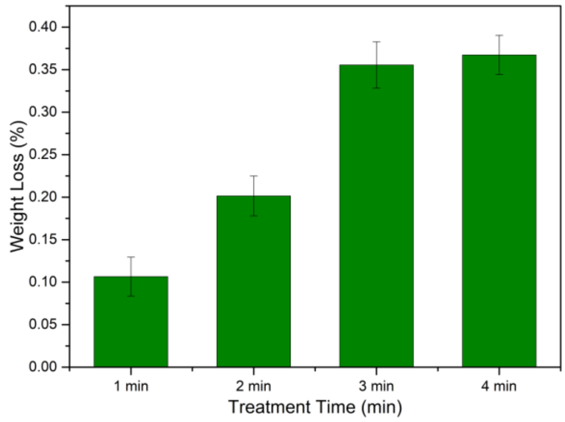

Figure 12: Weight Loss (%) at different treatment time

IV. Conclusions:

Atmospheric pressure 50 Hz DBD system successfully generated at Plasma Physics laboratory, Kathmandu University, Nepal was used for the surface treatment of Handmade Lokta paper to improve the hydrophilicity. The equivalent capacitance between the electrodes, the energy dissipated per cycle, and electron density in the plasma were measured by electrical methods and were found to be 8.3 pF, 3.54 mJ, and 8.47×10^{8} cm^{-3} respectively. The electron temperature in the plasma was found to be 1.29 eV. The results of wicking rate measurements confirmed that the exposure of plasma remarkably improved the hydrophilicity of the Lokta paper. The gravimetric analysis confirmed that there was a considerable weight loss (%) in the paper after the plasma treatment. These results concluded that 50 Hz atmospheric pressure DBD produced in air provide a simple, cost-effective, and reliable technology for the surface treatment of handmade Lokta paper. It can be used for long-term operations and continuous treatment of paper.

V. Data Availability

The authors can provide the data, figures, and articles that support the finding upon request.

VI. Conflicts of Interest

The authors declare no conflicts of interest.

VII. Acknowledgments

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VIII. Author Contributions

G.K.C. and R.P.G. designed the experimental setup; G.K.C., R.P.G., and D.P.S. performed the experiments; G.K.C., and R.P.G. analyzed the data; G.K.C. prepared the draft of the manuscript; R.P.G., U.M.J., and D.P.S. helped revise the manuscript.

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