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Atmospheric pressure plasma treatment of pure cotton textile for improved hydrophilicity and optimized dyeability via response surface methodology

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ABSTRACT

Raw cotton fibers contain non-cellulosic impurities that impart hydrophobic properties, making the dyeing process water- and energy-consuming. Conventional methods such as desizing, scouring, and bleaching involve hazardous chemicals, raising environmental concerns. In this study, the potential of atmospheric pressure plasma in surface modification to improve the hydrophilicity and dyeability of pure cotton textiles was investigated. Cotton samples were treated using an Atmospheric Pressure Plasma Jet (APPJ) system with an argon-oxygen gases mixture for 30, 60, and 90 seconds. Optical Emission Spectroscopy (OES) confirmed the presence of ionized argon and oxygen species in the plasma plume. Scanning Electron Microscope (SEM) images revealed surface etching, increased roughness, and microcrack formation after treatment. Increased absorbance of hydrophilic functional groups after plasma exposure were detected through a Fourier Transform Infrared Spectroscopy (FTIR). As plasma treatment time increased, significant improvements were observed in contact angle reduction, water absorbance time, and fabric weight indicating enhanced hydrophilicity. Moisture and dye uptake efficiencies also increased correspondingly. Optimization using the Box-Behnken Design of Response Surface Methodology identified the optimum conditions as 90 seconds of plasma exposure, 9 LPM oxygen flow, and 45 minutes dye contact time. The predicted dye uptake efficiency was 10.21%, closely aligning with the experimental value of 9.85% (3.56% relative error). Compared to untreated cotton (3.56%), plasma-treated samples achieved a 177% increase in dye uptake. These findings highlight the potential of APPJ treatment as a sustainable, energy-saving, and cost-effective approach for improving cotton textile processing.

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INTRODUCTION

Cotton (*Gossypium hirsutum* L.) represents the most profitable and significant non-edible crop globally providing a major source of livelihood for millions of farmers. It exhibits broad ecological adaptability to tropical and subtropical agroclimatic zones and is extensively cultivated for its high-value natural fibers (Mollaee et al. 2019). It is one of the most widely used natural fibers in the textile industry, valued for its comfort,

KEYWORDS

cotton, atmospheric pressure plasma, hydrophilicity, dyeability, response surface methodology

softness, breathability, and affordability (Lou et al. 2021; Patil and Netravali 2019). According to the latest report of the Philippine Fiber Industry Development Authority (PhilFIDA, 2024), the production and baling of commercial cotton fiber reached approximately 13 metric tons. Cotton accounts for approximately one-third of all fibers used in textiles in the Philippines, primarily due to its substantial production and its key role in common blended fabrics such as cotton-piña and cotton-abaca, both of which typically contain 70% cotton.

Cotton fibers consist of around 92% cellulose, and the rest is a complex mixture of non-cellulosic compounds, including wax, protein, pectin, and other substances. Although cellulose is inherently hydrophilic, these non-cellulosic components contribute to the limited water absorbency of raw cotton fibers (Colombi et al. 2021). Furthermore, during the weaving process, sizing agents commonly derived from starches like potato, corn, or rice are applied to cotton yarns to reduce friction, minimize breakage, and enhance weaving efficiency (Sandanuwan et al. 2021). While these agents improve the mechanical performance of the yarns, they further reduce the wettability of the finished fabric, presenting challenges in subsequent wet processing steps such as dyeing.

The non-cellulosic substances present in raw cotton, along with the sizing agents applied during the weaving process, interfere with the wettability of the fabric, thereby posing challenges in the dyeing and finishing procedures. Due to the low absorption and limited fastness properties of raw cotton textiles, the dyeing process becomes water-intensive and energy-consuming. Specifically, dyeing 1 kilogram of cotton fiber requires approximately 125 liters of water and substantial amount of energy for heating water and steaming. Moreover, approximately 200,000 tons of dye are wasted annually due to the inefficiency of dyeing cotton (Chequer et al. 2013). Cotton dyeing is a relatively lengthy procedure compared to synthetic fibers. While synthetic fibers typically undergo shorter processes and achieve 99% or more dye fixation, cotton can only take up about 50-70% of the dye used (Nallathambi and Rengaswami 2016; Fang et al. 2019)

The challenges regarding the dyeability of cotton fabrics also prompt a preference for synthetic dye over natural dye. Beyond their cost-efficiency and easy application, synthetic dyes offer a significantly broader spectrum of colors and produce more vibrant and stable colors compared to natural dyes. The synthetic dye industry, however, became one of the world's most polluting industries due to various chemicals and materials involved in its production, which are considered toxic and harmful to humans and the environment (Ticha et al. 2016; Sivakumar et al. 2011; Khattab et al. 2020)

Enhancing the water absorbency of cotton textiles is one of the most critical factors to ensure uniform coloring, achieve adequate fastness properties, and optimize dyeing efficiency. Thus, prior to dyeing and finishing, cotton textiles undergo pretreatment to remove impurities and achieve high purity of cellulose content, resulting in an improved hydrophilicity.

Wet chemical treatments such as desizing, bleaching, and scouring are the conventional methods used to prepare cotton textiles for further treatments (Kan et al. 2014). In desizing, the insoluble sizing agents that were applied prior to weaving are converted to water soluble compounds through physical or chemical processes or a combination of both. Bleaching involves the utilization of hydrogen peroxide (H_2O_2) to remove impurities, stains, and coloration from the fabric, enhancing the overall dye affinity of the material. Textile cleaning through scouring is conducted using alkaline chemicals, such as sodium hydroxide (NaOH), at boiling temperature, followed by multiple rinsing steps (Sivakumar et al. 2011; Kan et al. 2014). Although these wet chemical treatments are effective in enhancing the hydrophilic and dyeability properties of cotton textiles, drawbacks include high water and energy consumption, toxic byproducts from chemicals used at high temperatures, oxidation damage of the cellulose leading to reduced fiber strength, and environmental hazards from effluents. Thus, there is an increasing necessity in the textile industry to find alternative pretreatment methods that are cost-effective, energy-efficient, and environmentally friendly.

The application of plasma technology to textile materials has been gaining popularity due to its potential for surface modification, altering material performance without interfering with its bulk properties. Non-thermal plasmas are able to alter the physicochemical properties of polymers by introducing activated species, such as electrons, ions, and radicals, inducing new functional groups and modifying morphological properties. Some of the studied effects of plasma surface engineering are improved hydrophilicity, hydrophobicity, adhesion, and dyeability. Furthermore, non-thermal plasma modification is a relatively simple and dry method, therefore, no chemicals or wastewater are involved, hence, economic and environmental disadvantages can be eliminated. (Sandanuwan et al. 2021; Inbakumar et al. 2010; Nithya et al. 2011).

Over the past decade, numerous studies have demonstrated the effectiveness of non-thermal plasma in surface modification (Mohandoss et al. 2024; Vajpayee et al. 2021; Prado et al. 2016). However, limited research has explored its potential as a viable alternative to conventional chemical pretreatments for improving the hydrophilicity and dyeability of cotton textiles. In particular, there is a scarcity of studies addressing the optimization of working parameters for atmospheric pressure plasma jet (APPJ) systems in textile surface modification. Traditionally, parameter optimization has been conducted using the one-factor-at-a-time approach, where individual variables are altered while others are held constant. While straightforward, this method is time-consuming, resource-intensive, and often fails to capture the interactions between variables.

A more efficient and systematic approach is the use of Response Surface Methodology (RSM), a robust statistical technique that integrates experimental design, process optimization, and modeling for multiple variables simultaneously. Among the various RSM designs, the Box-Behnken Design (BBD) is widely adopted due to its efficiency and reduced number of required experimental runs. BBD enables the identification of optimal process conditions with minimal resource consumption while providing reliable insights into the relationships among variables (Haji 2017; Bulatao et al. 2019).

The present study investigated the potential of atmospheric pressure plasma (APP) treatment to enhance the hydrophilicity and dyeability of pure cotton textiles through surface modification. Additionally, the dyeing process was optimized using the Box-Behnken Design of Response Surface Methodology (RSM).

MATERIALS AND METHODS

Atmospheric Pressure Plasma Jet Setup

The modified Atmospheric Pressure Plasma Jet (APPJ) system used in this study is shown in Figure 1 (Malapit and Baculi 2022). It features a quartz glass chamber equipped with two gas inlets, two ports for rubber stoppers securing the electrodes, and a 2mm nozzle serving as the plasma plume outlet. Argon and

oxygen gases (99.9% purity) are introduced as feed gases and ignited via a pair of 1.50 mm diameter copper electrodes (99.50% purity), positioned 5 mm apart. These electrodes are powered by a 60 Hz, 450 W neon sign transformer delivering 15 kV and 30 mA. A variable autotransformer (variac) is connected to the neon transformer to allow precise adjustment of the output voltage.

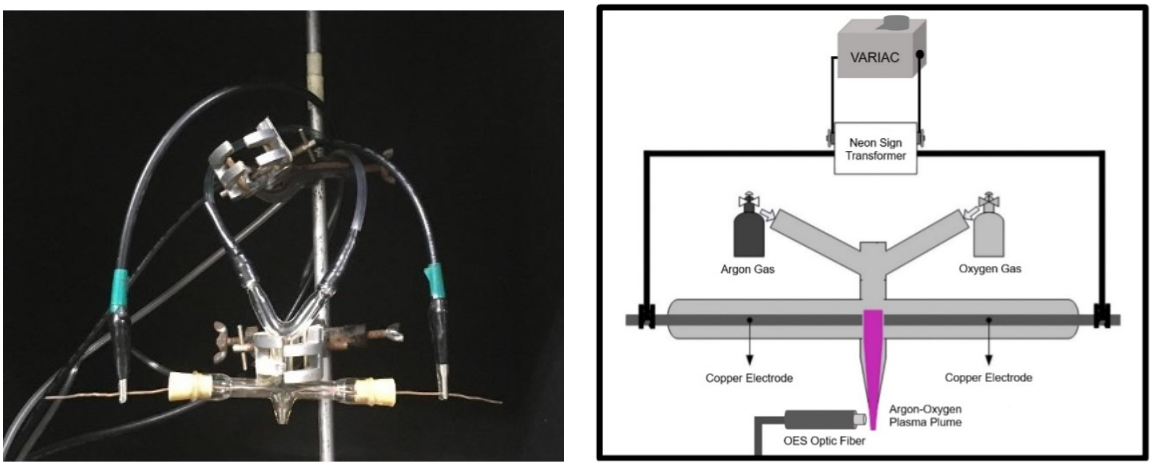


Figure 1: Experimental setup of atmospheric pressure plasma jet (APPJ) (left) and a schematic diagram of the APPJ system (right)

Sample Processing

The traditional pure cotton textile was obtained from the Cordillera Textiles Project (CordiTex) of UP Baguio. The textile was cut into 1.50 by 1.50 cm samples prior to plasma exposure. The samples were treated with non-thermal plasma generated using argon and oxygen gases with 2.50 LPM and 6 LPM flow rates, respectively. From the specified flow rates, the total volumetric flow rate was approximately 1.42×10^{-4} m³/s, and the corresponding plume velocity, calculated using the continuity equation, was 45.2 m/s. The output voltage of the system was adjusted to approximately 7.80 kV (variac set at 120V). The corresponding output voltage to a given variac input can be estimated using the linear equation:

$$y = 64.716x - 10.458$$

Where:

X is the variac input voltage.

This relationship was established through linear regression analysis of experimental data correlating variac settings with measured output voltages.

The nozzle to substrate distance was kept constant at 1 cm and treated with increasing plasma treatment time: 30, 60, and 90 seconds. Since the APPJ setup available only covers 0.50 by 0.50 cm of the sample's surface, the samples were manually raster-scanned in vertical and then horizontal directions, alternatingly. This was done to ensure uniform plasma exposure across the entire substrate surface and to prevent localized overheating or damage to the textile sample.

Plasma Discharge Characterization

The dominant species present in the generated plasma was verified using an Optical Emission Spectrometer. A fiber optic cable connected to the spectrometer was pointed towards the plasma plume. The spectrometer was connected to a computer which records the spectral lines emitted by the plasma discharge through the OceanView software. The captured emission spectra data was then analyzed using the Spectrum Analyzer 1.97 software. Furthermore, the same software was used to determine the electron temperature of the plasma discharge. In order to verify that the generated plasma is non-thermal or "cold", the discharge temperature was measured using a Cooper-Atkin's K thermocouple.

Cotton Surface Characterizations

The cotton samples, measuring 1.5 by 1.5 cm, were subjected to plasma treatments of 30, 60, and 90 seconds. To facilitate easier identification, the treated side of the cotton textiles were marked, since only one side of each sample underwent plasma treatment. They were then stored in glass petri dishes, labeled, and subsequently subjected to physical and chemical surface characterization.

The surface morphology of the untreated and plasma-treated cotton textiles was analyzed using the Hitachi TM4000Plus SEM at the Philippine Science High School Ilocos Region Campus (PSHS IRC). Cotton samples were subjected to SEM imaging without any prior conductive coating. The analysis was employed under the following conditions: resolution: 1 mm – 50 µm; magnification: 30x – 800x; accelerating voltage: 20 kV. The resulting magnified images of untreated and treated cotton samples were then compared to observe the physical changes that occurred after plasma treatment.

A PerkinElmer Spectrum Two FTIR Spectrometer with a Diamond Attenuated Total Reflectance (ATR) from the PSHS IRC Laboratory was used to analyze the functional groups of the untreated and plasma-treated textile surfaces. Spectra were collected in the range of 4000 to 700 cm^{-1}. Data below 700 cm^{-1} were excluded due to significant instrument noise and a low signal-to-noise ratio. Peaks corresponding to different functional groups were then identified and interpreted from the resulting transmission and absorbance graphs.

Cotton Surface Hydrophilicity Tests

The hydrophilicity of the cotton surface was assessed using Contact Angle Measurement, Water Absorbency Test, Moisture Uptake Test, and Weight Loss Test.

Contact Angle Measurement

The untreated and plasma-treated cotton samples were subjected to a sessile drop method to measure the water contact angle and assess their wettability. Using a micropipette, 50 µl of distilled water was dispensed onto the sample. The image of the droplet on the textile surface was captured using a digital microscope connected to a computer. The images were then analyzed using the DropSnake plugin (Stadler et al. 2010) feature of the ImageJ (Schneider et al. 2012) software to determine the shape and calculate the contact angle of the water droplets. To assure the reliability of the experiment, three replicates for each treatment were used.

Water Absorbency Test

The measurement of water absorbency for the textile samples was adopted from the test methods of The Textile Institute (Saville 1999). A 50 µl droplet of water is released from a pipette to the

horizontal surface of the textile sample, illuminated at a 45° angle. The time it took for the diffuse reflection from the liquid to vanish is measured and recorded.

Moisture Uptake Test

In accordance with the standard procedure indicated in ISO 20158:2018, the untreated and plasma-treated textile samples were conditioned in a standard atmosphere with a relative humidity of 65 ± 2% for 24 hours prior to initial weighing. Cotton samples were then submerged in water for 120 ± 2 seconds and left to dry for 60 ± 2 seconds. Afterwards, the samples were weighed using an analytical balance to determine the final weight to the nearest 0.01 grams.

The moisture uptake or the water absorption capacity (WAC) of the samples were calculated using the equation

$$WAC\ (\%) = \left(\frac{m_2 - m_1}{m_1}\right) x\ 100$$

Where:

m_1 is the mass (in grams) of the cotton textile sample in dry state; and

m_2 is the mass (in grams) of the cotton textile sample in wet state.

Weight Loss Test

For the weight loss test, untreated samples were conditioned for 24 hours at standard atmospheric condition with a relative humidity of 65 ± 2% before the initial weights were measured. The final weight was measured after subjecting the samples to different durations of plasma treatment: 0s, 30s, 60s, 90s. The weight loss percentage was calculated using the formula:

$$Weight\ loss\ (\%) = \left(\frac{w_1 - w_2}{w_1}\right) x\ 100$$

Where:

w_1 is the mass (in grams) of the cotton textile sample prior to plasma treatment; and

w_2 is the mass (in grams) of the cotton textile sample after plasma treatment.

Dye Uptake Efficiency Measurement

A 20 g/L dye stock solution was prepared by dissolving 2 g of commercially available synthetic azo dye powder (Master Fast Dyes by Colorscape Inc., Philippines) in enough distilled water to make a 100 mL solution. It was then diluted to achieve a 0.2 g/L working solution. Untreated and treated cotton samples were submerged in 6 mL of diluted working dye solution and subsequently removed from the dye bath solution after 15, 30, and 45 minutes, in accordance with the parameters generated by the software.

In order to measure the dye uptake of the samples, the absorbance of the used dye bath solution was measured through a UV-Vis available at the Chemistry Laboratory of UP Baguio.

From the resulting absorbances of the dye bath solutions used, the dye uptake efficiency was calculated using equation

$$Dye\ uptake\ efficiency\ (\%) = \left(\frac{d_1 - d_2}{d_1}\right) x\ 100$$

Where:

d_1 is the concentration of the original dye used; and

d_2 is the concentration of the dye bath used to saturate the cotton samples.

Here, absorbance values were used in place of concentrations to calculate the percent dye uptake efficiency. The main component of the commercially available azo dye was not specified in the product details, and thus its molar absorptivity is unknown. While Beer's Law typically requires the molar absorptivity to convert absorbance to concentration, it is not necessary in this case because the same constant applies to both the initial and final measurements. As a result, it cancels out in the calculation, allowing absorbance values to yield an accurate percent dye uptake efficiency.

Dyeability Optimization via Response Surface Methodology (RSM)

The Stat-Ease 360 software was used to create the experimental design and statistical evaluation of responses. RSM was employed to find the optimal plasma treatment parameters and dyeing conditions to attain maximum efficiency of dye uptake. In this study, RSM was conducted in three stages: (1) screening, (2) optimization, and (3) verification.

In the screening process, a two-level full factorial design was used to determine the significance of the chosen parameters and their interactions. Test runs were produced randomly by the software using the experimental factors chosen for this study: plasma treatment time, oxygen gas flow rate, and dye bath exposure time. Table 1 shows the factors and the values used for the screening stage.

Table 1: Experimental ranges of factors for the two-level full factorial design

Factor	Name	Unit	Low level (-1)	High Level (+1)
A	Plasma treatment time	Seconds	30	90
B	Oxygen gas flow rate	LPM	3	9
C	Dye bath exposure	Minutes	15	45

Box-Behnken Design (BBD) was employed to evaluate the effect of the three experimental factors on the dye uptake efficiency in order to provide the optimal combination of plasma and dyeing conditions for the cotton textile samples. The factors and values used for the optimization stage are shown in Table 2, where the three levels -1, 0, 1, represent the low, middle, and high levels, respectively. Triplicates of the software-produced runs, except for the center points, were performed to ensure the reliability of the experiment.

Table 2: Experimental ranges of factors for the Box-Behnken design

Factor	Name	Unit	-1	0	1
A	Plasma treatment time	Seconds	30	60	90
B	Oxygen gas flow rate	LPM	3	9	15
C	Dye bath exposure	Minutes	15	30	45

Experimental data were fitted to the second order regression and the significance of the model was determined using Analysis of Variance (ANOVA). Verification of the model was determined by obtaining experimental results using the given optimized parameters, which was then compared to the predicted maximum response value generated by the software.

RESULTS AND DISCUSSION

Plasma Generation and Characterization

The mixture of argon and oxygen gas, ignited between two electrodes with an applied voltage of 7.80 kV, generated a turbulent flow of light purple plasma discharge. Argon plasma produces a violet glow, while oxygen plasma has a faint white/gray color hence, the argon-oxygen plasma produced has a pale purple

discharge due to the higher oxygen content in the gas mixture. This is consistent with the characteristics of argon/oxygen gas mixtures in atmospheric plasma, as studied by the works of Fang and his colleague (Fang et al. 2016)

Optical Emission Spectroscopy was employed to determine the species present in the generated plasma. Figure 2 shows the wavelength-intensity graph, as well as the identified peaks, of the optical emission spectrum data. In order to yield straightforward results, the Spectrum Analyzer software was configured to exclusively detect the presence of argon, oxygen, and copper ions.

The presence of neutral argon species (Ar I) was detected at 810.37 nm. The species of neutral oxygen (O I) was located at 799.51 nm. On the other hand, peaks of singly-ionized argon atoms (Ar II) were identified at 761.80 nm and 839.57 nm. Singly-ionized oxygen species (O II) were observed at 575.39 nm, 736.49 nm, and 748.63 nm. Lastly, singly-ionized copper atoms (Cu II) appeared at 695.29 nm and 775.44 nm.

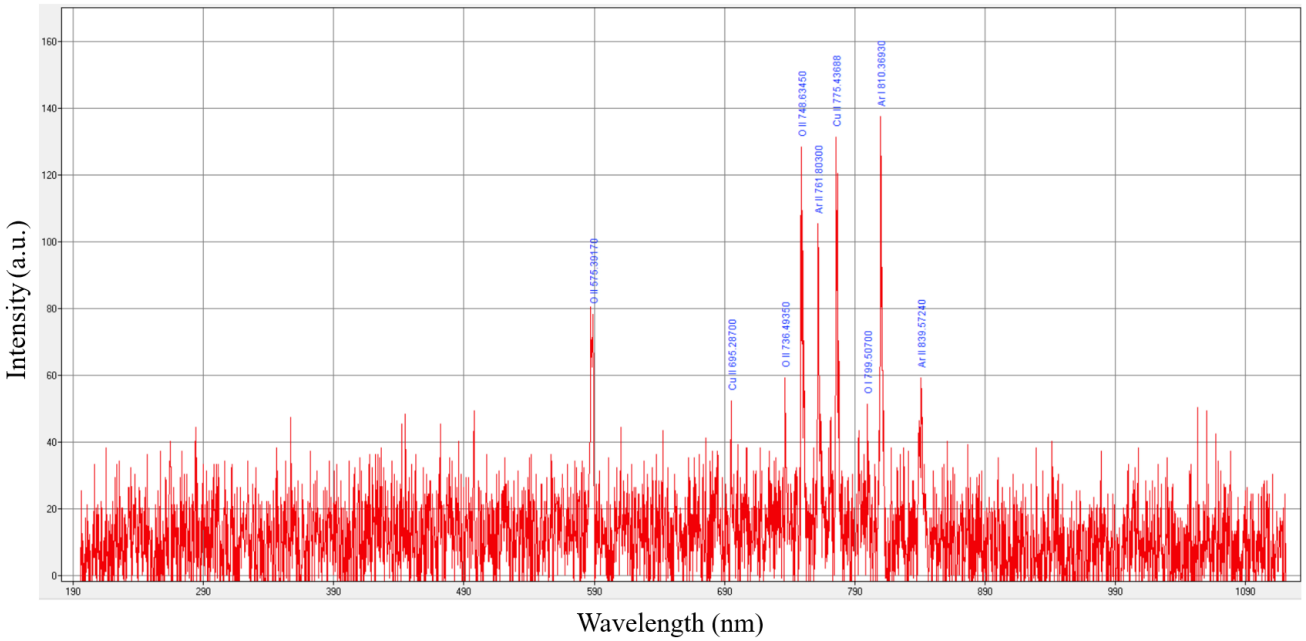


Figure 2: Optical emission spectrum of the atmospheric pressure plasma discharge

The identification of ionized argon and oxygen atoms in the spectrum data implies that the discharge generated contained ionized gas, further confirming that the discharge was an argon-oxygen plasma. To be classified as "cold" plasma, the gas temperature should be significantly lower than the electron temperature.

Using the Spectrum Analyzer 1.97 software, the excitation temperature of the plasma was measured to be 8,444 K (~ 0.73eV), with a correlation coefficient of r = 0.97. This was determined using intensity values of Ar I spectral lines and the Boltzmann regression method represented by the equation:

$$ln\frac{I_{kl}\lambda_{kl}}{g_{kl}A_{kl}} = \frac{E_k}{k_B T_e} + C$$

Where:
I_{kl} is the intensity of the emitting light
λ_{kl} is the wavelength
g_{kl} is the statistical weight of upper level
A_{kl} is the value for the transition probability from level k to level l
E_k is the excitation energy
k_B is the Boltzmann's constant
C is a constant.

This approach is adapted from the method proposed by Moon and Choe (2006), which found that the excitation temperature exhibits a similar trend to the electron temperature in APPJ setups. In addition, Wali et al. (2024) report that the electron temperature of Ar-O_2 plasma increases with the addition of O_2 due to the generation of rotational and vibrational excited states via electron collisions.

On the other hand, the measured average gas temperature of the plasma discharge is 432K. Since the condition is satisfied, the generated discharge is then classified as non-thermal or cold plasma.

Cotton Surface Characterizations

SEM was used to observe the surface of raw and plasma treated cotton textile samples. Figure 3 shows the SEM images of the cotton samples at 30x magnification. Results show that the plasma treatment caused etching of the hair-like fiber of the cotton fabric. This is most evident for the cotton sample with the highest plasma treatment time applied (90 s), as highlighted by the yellow circles.

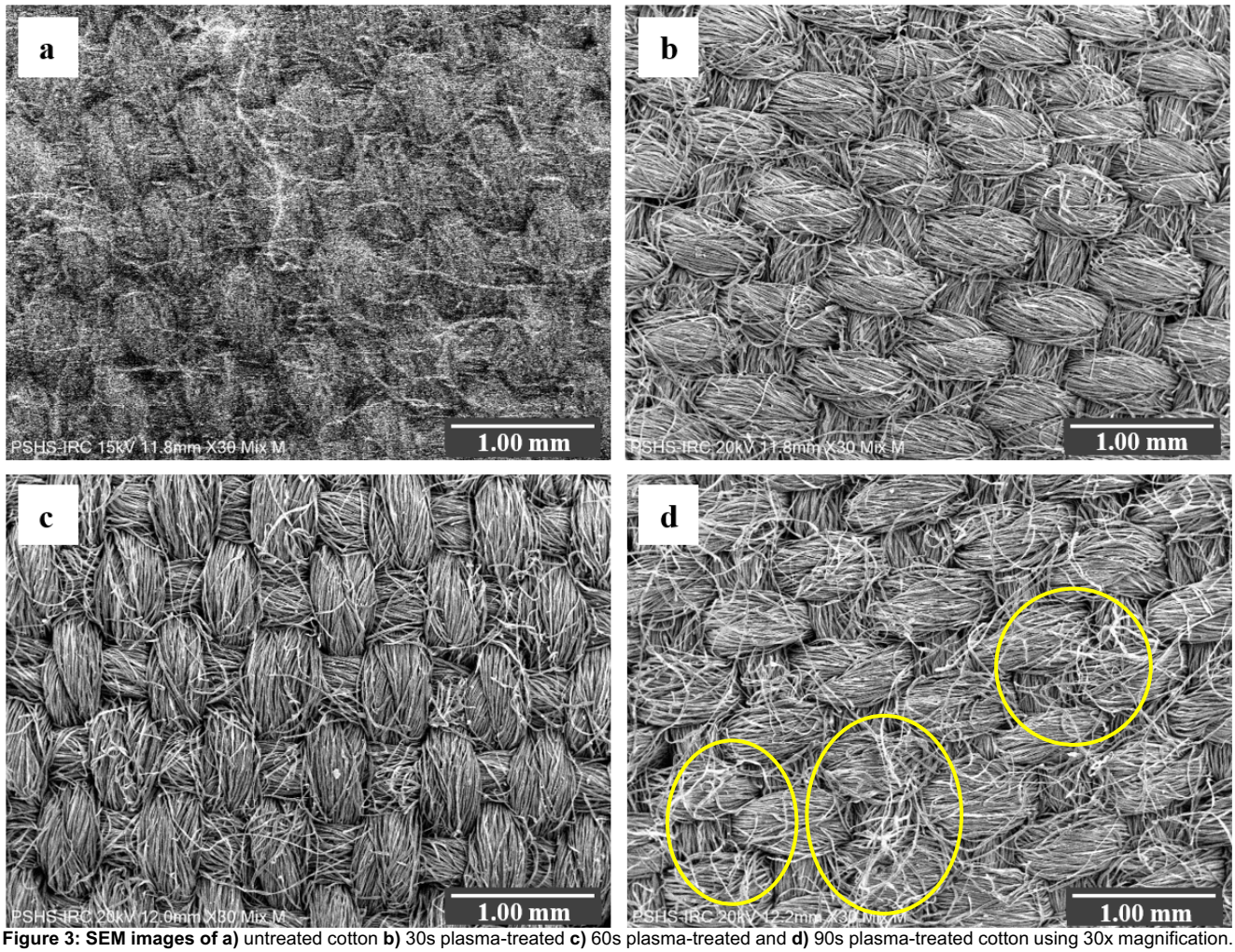


Figure 3: SEM images of a) untreated cotton **b)** 30s plasma-treated **c)** 60s plasma-treated and **d)** 90s plasma-treated cotton using 30x magnification. The areas marked with yellow circles in the image indicate the most prominent etching observed on the hair-like fibers of the cotton samples

Typically, sizing agents are observed on raw cotton fibers, as they play a vital part in the weaving process of cotton textiles. Cotton fibers also contain non-cellulosic materials, such as pectins, waxes, proteins, and sugars, that reside on the cuticle layer of the fiber (Nithya et al. 2011). These impurities on the outermost layer of the cotton fiber, which are hydrophobic in nature, are mainly responsible for the poor ability of raw cotton textiles to absorb water. Studies show that oxygen plasma has applications in surface "cleaning" of textiles achieved through the etching of sample surfaces (Peran and Ercegovic Razic 2020). Verifying the presence and the removal of impurities on the surface of raw cotton samples, caused by etching induced by plasma treatment, using only SEM images can be challenging. It can be, however, presumed that plasma treatment can potentially remove the waxes and non-cellulosic materials from the surface of the raw cotton samples using the results from the FTIR analysis, contact angle measurement, and weight loss test, which are discussed in the following sections.

Further observations were conducted using 600x – 800x magnification settings of the SEM, resulting images are shown in Figure 4. Raw cotton samples displayed smooth fibers, while plasma affected the roughness of the treated samples. The emergence of microcracks and grooves on the surface of the treated samples can be attributed to the direct bombardment of active species present in the plasma (Kan et al. 2014). The increase in surface roughness and the appearance of etched fibers provide new pathways for liquid to enter and interact with the treated samples, thus enhancing hydrophilicity. On the other hand, bombardment of oxygen species may improve the adhesion of the sample surface, consequently enhancing its dyeability properties (Pandiyaraj and Selvarajan 2008).

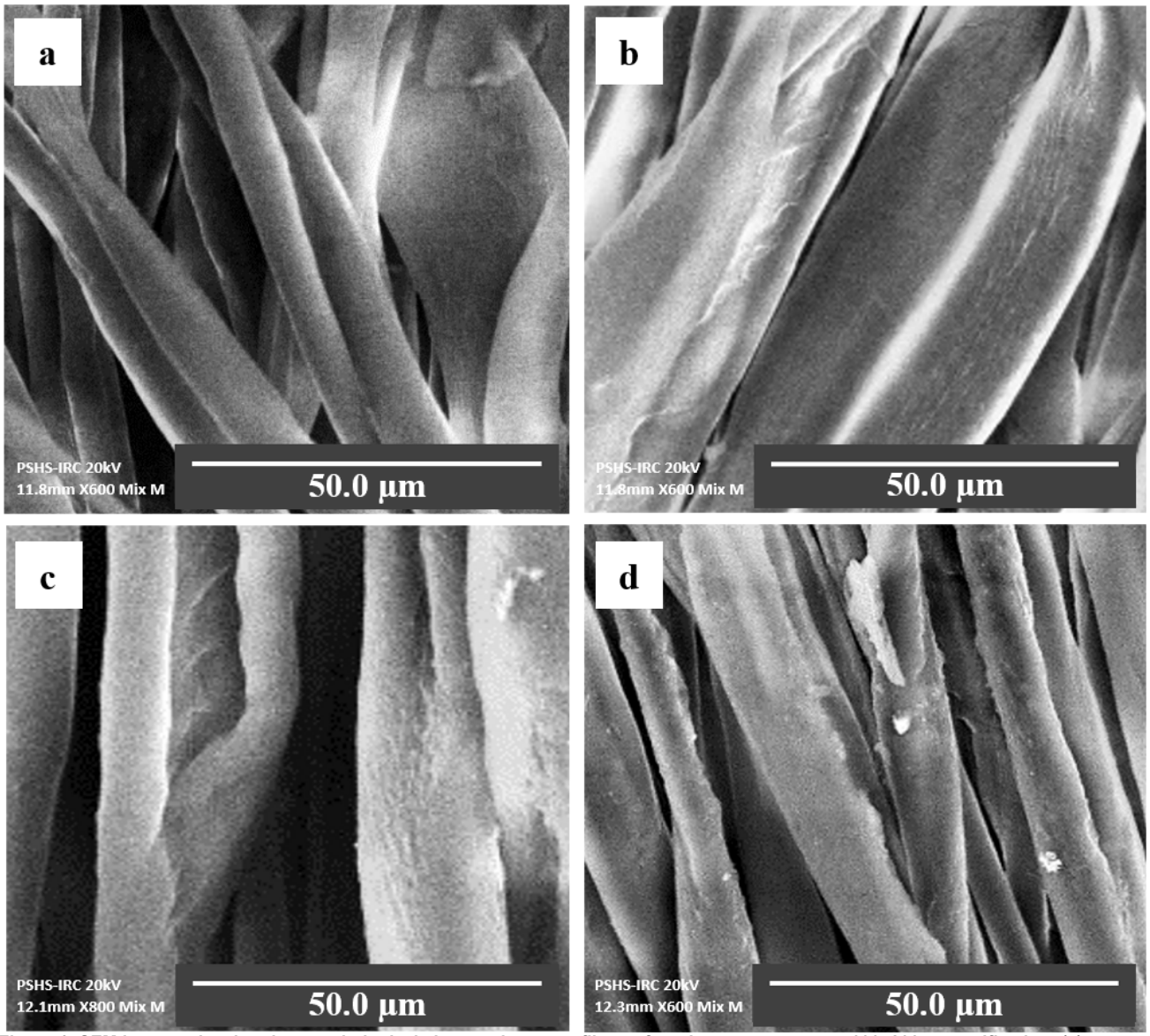


Figure 4: SEM images showing the morphological changes in cotton fibers after plasma treatment at 600–800× magnification: (a) untreated cotton, **(b)** 30 s plasma-treated, **(c)** 60 s plasma-treated, and **(d)** 90 s plasma-treated.

On the other hand, untreated and 30s plasma-treated cotton textiles were subjected to FTIR-ATR analysis to study changes in surface chemistry and determine the functional groups present in the samples. The resulting infrared spectrum of untreated and treated cotton, represented by the transmission data, is reported in Figure 5. Major peaks are observed at 3333 cm^{-1}, 2901 cm^{-1}, 1632 cm^{-1}, and 1053 cm^{-1}.

It can be noted that the absorption intensity is proportional to the concentration of the corresponding functional group (Pandiyaraj and Selvarajan 2008) and absorbance is calculated as a logarithmic function of transmittance. Hence, an inverse relationship is observed in the resulting values, that is, a decrease in transmittance percentage ensues an increase in the absorbance value.

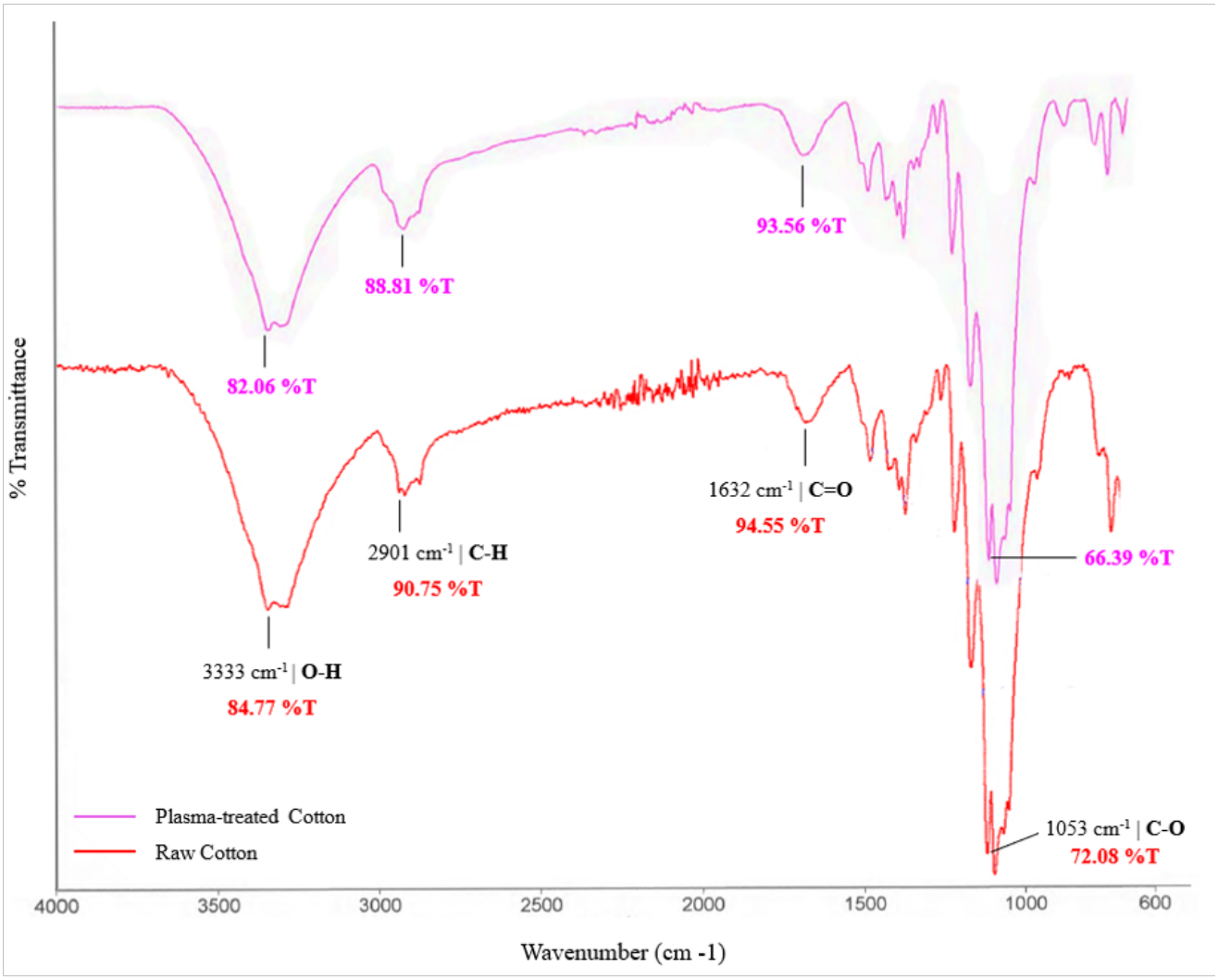


Figure 5: FTIR spectra of untreated and 30-s plasma-treated cotton

The peak analysis for this study was paralleled to the works of Caschera et al. (2014) and Kan et al. (2014) on the wettability and dyeability of cotton fabrics (Kan et al. 2014; Caschera et al. 2014). The spectra obtained from the untreated and treated cotton textiles are similar, however, a difference in peak intensities is observed after subjecting raw cotton samples to argon-oxygen plasma. Peaks at the 3200 – 3400 cm^{-1} region represent O-H stretching, indicating the presence of hydroxyl groups. C-H stretching peaks are observed around 2800 – 2980 cm^{-1} and C-O stretching occurs at 1053 cm^{-1}. The peaks recognized in these regions can be associated with the cellulose structure or the typical backbone of cotton fibers (Kan et al. 2014). It is observed that peaks in these regions decreased after plasma treatment, which can indicate an increase in the detected purity of cellulose.

The raw cotton samples typically contain non-cellulosic materials on the outer layer, which appear likely to be removed through plasma etching, as seen from the SEM images. Thus, the increase in peaks associated with the cellulose structure of cotton may be attributed to an increased exposure of cellulose material caused by the etching of the outermost layer or cuticle after plasma treatment.

The sharp peak located at 1053 cm^{-1}, and stretching along 1657 – 1605 are associated with oxygen containing groups, specifically, C-O and C=O stretching. The transmission data for the plasma-treated cotton within these regions decreased in intensity, implying an increase in oxygen functional groups. The bombardment of oxygen plasma on the cotton textile surface resulted in the incorporation of oxygen species into the cotton sample's structure, forming oxygen-containing polar groups. This has a significant effect on the enhanced hydrophilicity of the material (Caschera et al. 2014).

Generally, highly reactive but short-lived oxygen species such as OH radicals, NO, and O_2^- are generated through excitation, ionization, and recombination processes in the plasma. These species exist only for microseconds and decay within a few millimeters from the plume origin. Due to their short lifetimes, they rapidly undergo secondary reactions with neutral molecules or water vapor in the ambient environment, forming longer-lived reactive oxygen species (ROS) such as hydrogen peroxide (H_2O_2) and ozone (O_3). These longer-lived species can persist for milliseconds to several days, allowing them to travel further and interact with substrates located at greater distances from the plasma source (Adhikari et al. 2021). Therefore, it can be inferred that long-lived ROS, particularly H_2O_2 and O_3, were primarily responsible for the effective functionalization of the plasma-treated sample.

Hydrophilicity Evaluation

Contact Angle

The interaction of water droplets on the surface of untreated and plasma-treated cotton samples are shown in Figure 6, and the decreasing trend of average contact angle measurement as plasma exposure time was increased is shown in Figure 7. Raw cotton textiles were measured to have an average contact angle of 143.41 ± 0.93°. The average contact angle decreased to 52.96 ± 0.49° after 30 seconds of plasma exposure, and continued to decline to an

average of 21.34 ± 1.35° after 60 seconds of plasma treatment. Cotton samples that were treated with plasma for 90 seconds exhibited complete wetting behavior, as evidenced by the water droplet spreading across the surface within the first few seconds of contact, yielding approximately 0° contact angle.

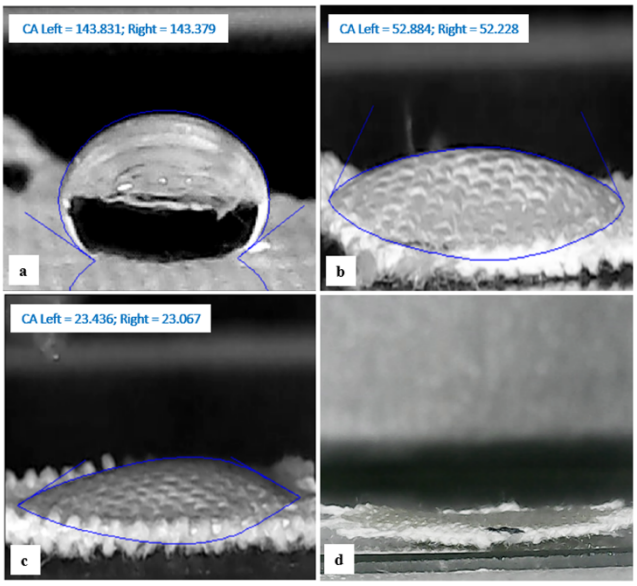


Figure 6: Contact angle measurement of a) untreated **b)** 30s plasma-treated **c)** 60s plasma-treated and **d)** 90s plasma-treated cotton textiles

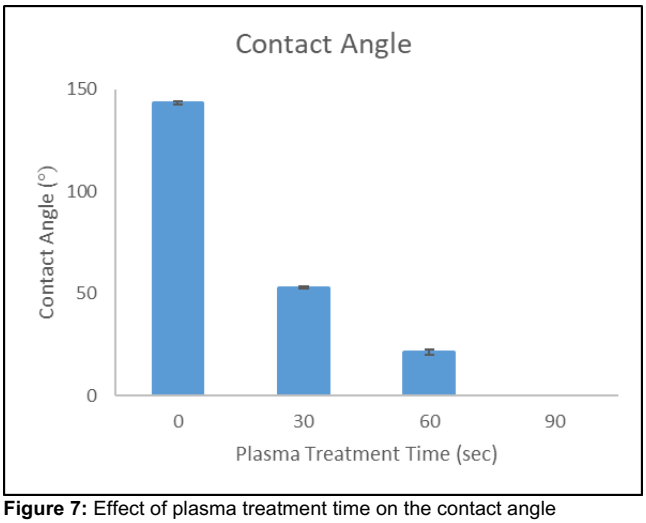


Figure 7: Effect of plasma treatment time on the contact angle

The contact angle represents the measured angle formed between a solid surface and the tangent to the water surface as it approaches the solid. A material with a contact angle greater than 90° is considered hydrophobic, while a contact angle less than 90° implies hydrophilic properties (Saville 1999).

Hydrophilicity is associated with a low contact angle measurement, signifying the material's affinity for water absorption, whereas materials with a high contact angle tend to repel water, causing it to flow away from the surface (Saville 1999). As shown in Figure 3.6, the contact angle between the water droplet and the cotton samples decreased with prolonged plasma exposure time. This can be attributed to the increased surface roughness of the cotton post-plasma treatment, as verified by the SEM results. The introduction of absorption sites facilitated water penetration among the protrusions, enabling water to move across the surface and thereby diminishing the contact angle.

Water Absorbency and Moisture Uptake

The Textile Institute defines wettability as the duration (in seconds) for a water droplet to permeate the fabric. Materials exhibiting a water droplet absorption time exceeding 200 seconds are classified as unwettable or hydrophobic. Table 3 summarizes the result of the water absorbency test, revealing that untreated cotton textiles exhibited the longest water absorbance time, exceeding 200 seconds. A significant decrease in absorbance time was evident following plasma treatment, where the 90-second treatment time obtained the shortest average wetting time at 2.293 ± 0.046 seconds.

Table 3: Effect of plasma treatment time on water absorbency of the cotton textiles

	WATER ABSORBANCE RATE (sec)				
Plasma Treatment Time (s)	***Trial 1***	***Trial 2***	***Trial 3***	***Average***	***Standard Deviation***
0	> 200	> 200	> 200	**> 200**	0
30	75.26	66.82	68.89	**70.32**	3.59
60	37.44	30.22	21.33	**29.66**	6.59
90	2.34	2.31	2.23	**2.29**	0.05

The efficiency of plasma-treated cotton samples to take up water was evaluated using the moisture uptake test, results are reported in Figure 8. Subsequent to 120 seconds of water saturation, the raw cotton fabric was measured to have an average of 18 ± 5% water absorbance capacity (WAC). The samples exposed to 30, 60, and 90 seconds of plasma treatment were observed to have elevated average WAC of 175 ± 10%, 179 ± 6%, and 197 ± 9%, respectively.

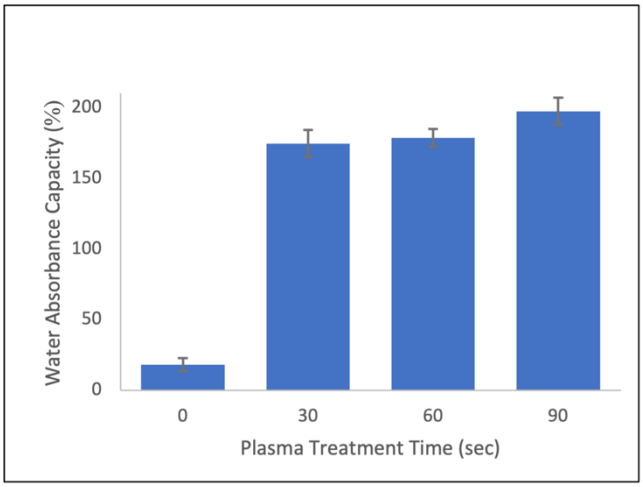


Figure 8: Effect of plasma treatment time on the water absorbance capacity of cotton textiles

An appropriate combination of surface chemistry and surface roughness is associated with the achieving hydrophobic and hydrophilic properties of fabrics. Therefore, the observed increase in absorbance of hydrophilic functional groups on the plasma-treated samples, along with the surface roughness induced by etching, may account for the rapid wetting and increased water absorbance capacity of these samples compared to the raw cotton samples.

Weight Loss

The average weight loss percentage of the cotton samples with and without plasma treatment are shown in Figure 9. Untreated cotton samples lost an average of 0.59 ± 0.11 % from their initial weight, which may be due to the conditioning environment of the samples. On the other hand, an upward trend in weight loss was observed along with increasing plasma exposure time. A 30-second plasma treatment resulted in an average of 4.36 ± 0. 06% weight loss. After 60 seconds of plasma exposure, the average weight loss percentage was calculated to be 4.58 ± 0.57%. Lastly, the weight loss efficiency rose to 5.51 ±0.80% after 90 seconds of plasma

exposure. The results of the weight loss test suggest that the etching induced by plasma treatment removed the wax, sizing agents, and non-cellulosic materials from the surface of raw cotton textiles.

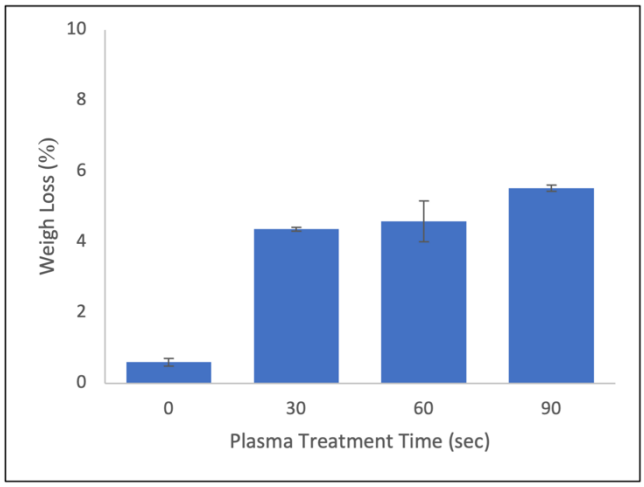


Figure 9: Effect of plasma treatment time on % weight loss of cotton textiles

Dye Uptake Evaluation and Optimization

Screening via Two Level Full Factorial Design

A total of 29 runs, including 5 center points and triplicates of 8 combinations of parameters, were randomly produced using the two-factor full factorial design. Table 4 shows the software-generated experimental design employed in the screening stage to evaluate the significance of the experimental factors: plasma treatment time, oxygen gas flow rate, and dye bath exposure. The corresponding response, representing dye uptake efficiency, is also shown.

Table 4: Experimental factors and responses for the two-level full factorial design

	Factor 1	Factor 2	Factor 3	Response
Run	**Plasma Treatment Time (seconds)**	**Oxygen Flow Rate (LPM)**	**Dye Bath Exposure (minutes)**	**Dye Uptake Efficiency (%)**
1	30	9	45	8.16
2	90	3	45	9.09
3	30	3	45	7.53
4	90	3	45	9.71
5	60	6	30	8.23
6	90	9	45	9.86
7	30	3	15	4.36
8	90	9	45	9.99
9	30	9	15	5.70
10	90	9	15	7.82
11	90	3	45	9.32
12	30	9	45	8.60
13	30	3	45	7.31
14	30	9	15	6.69
15	30	3	15	5.39
16	90	3	15	6.42
17	90	9	15	7.27
18	30	3	15	6.25
19	60	6	30	8.88
20	60	6	30	8.88
21	30	9	45	8.82
22	90	9	45	9.24
23	90	3	15	7.33
24	90	3	15	8.16
25	30	9	15	6.17
26	30	3	45	7.15
27	60	6	30	8.26
28	90	9	15	7.90
29	60	6	30	8.80

Table 5 presents the Analysis of Variance (ANOVA) results of the responses from the experimental model, where a p-value less than 0.05 indicates significance at the 95% confidence level and p-values greater than 0.1 are considered insignificant. The model F-

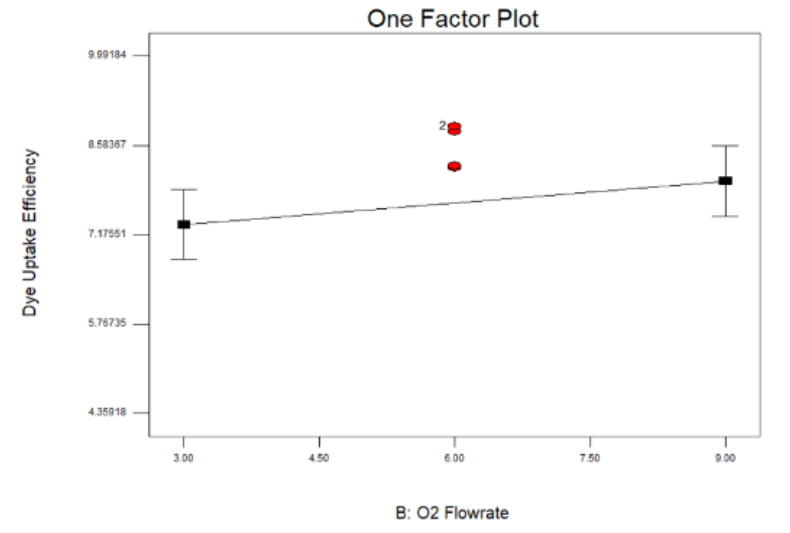
value of 30.55 and p-value of < 0.001 implies that the established model is significant. Moreover, the significance of model terms A, B, and C is apparent based on the obtained p-values. This implies that all experimental factors used (plasma treatment time, oxygen flow rate, and dye bath exposure) significantly affects the dye uptake efficiency. This is further validated by the positive graphs generated for each factor, as shown in Figure 10.

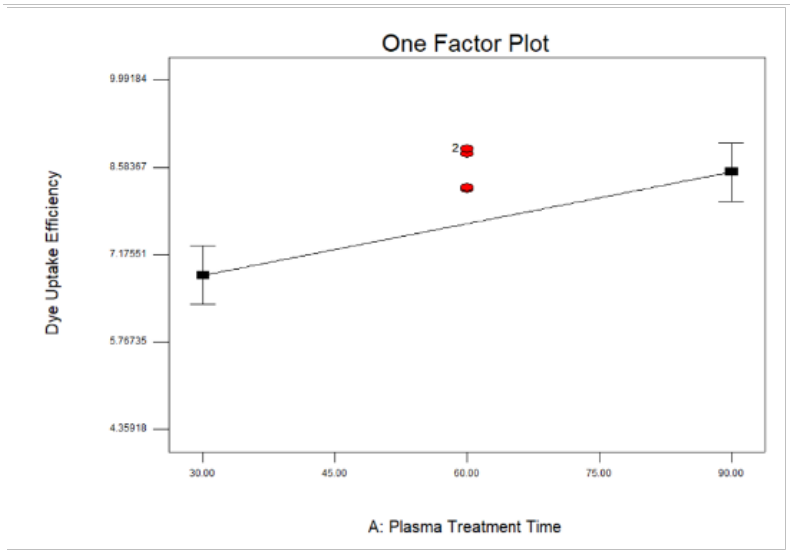
The curvature, having a p-value of 0.0012, was also found to be significant, indicating that at least one variable was involved in the higher order interaction. Since the curvature is indicative of a non-linear relationship between a factor and the response variable, it suggests that the optimum region would be near or within the experimental ranges of the independent variables (Bulatao et al. 2019). This aligns with the significance of the B^2 model term, as discussed in the ANOVA results in the following section.

Table 5: ANOVA results of the two-level full factorial design.

Source	Sum of Squares	DF	Mean Square	F-value	p-value Prob > F
Model	46.95	6	7.83	30.55	< 0.0001
A	16.64	1	16.64	64.97	< 0.0001
B	2.80	1	2.80	10.93	0.0034
C	26.75	1	26.75	104.46	< 0.0001
AB	0.71	1	0.71	2.75	0.1119
AC	0.020	1	0.020	0.076	0.7848
BC	0.035	1	0.035	0.14	0.7160
Curvature	3.60	1	3.60	14.07	0.0012
Residual	5.38	21	0.26		
Lack of Fit	0.054	1	0.054	0.20	0.6560
Pure Error	5.32	20	0.27		
Cor Total	55.93	28			

*p-value <0.05 indicates significance





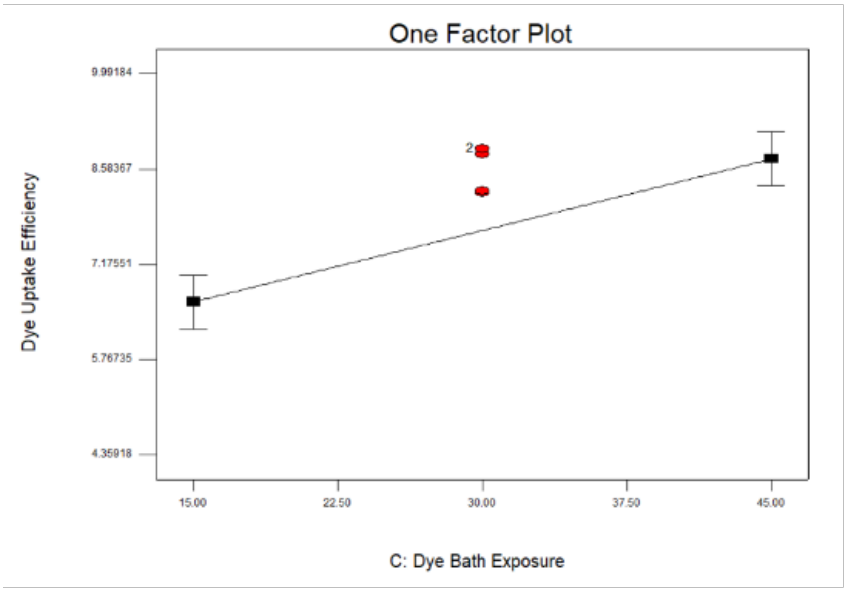


Figure 10: Effects of a) plasma treatment time **b)** oxygen flow rate and **c)** dye bath exposure on the dye uptake efficiency. Points on the center are center points

To highlight the differences in color intensity between untreated and plasma-treated cotton textiles, samples were submerged in the undiluted dye stock solution. It is evident from Figure 11 that the untreated cotton textile exhibited poor dyeability. This is mainly due to the hydrophobic impurities on the outermost layer of the cotton. As previously discussed, plasma treatment presumably removed these impurities, and subsequent etching led to an increase in the surface roughness of the sample. This increase in surface roughness enhanced the adhesion ability of the sample, hence, more dye particles were absorbed compared to raw cotton. Furthermore, the increased hydrophilicity, induced by the chemical composition changes on the plasma-treated surface, contributed to the improvement of the dye uptake capacity of cotton. This observation further supports the positive correlation between plasma treatment time and dye uptake efficiency, as illustrated in Figure 10a.

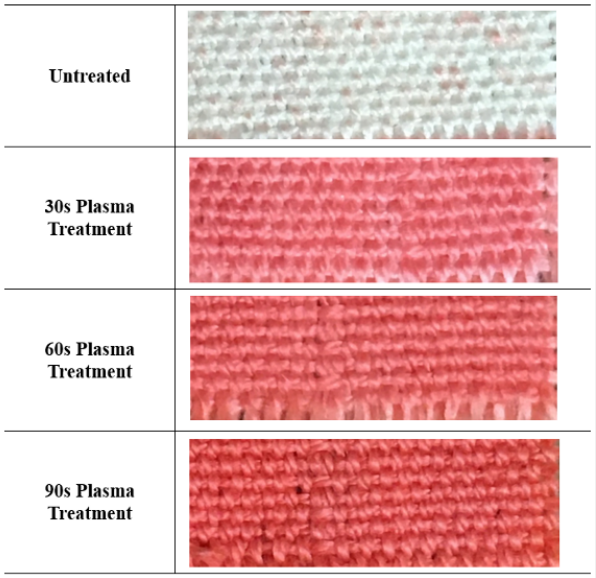


Figure 11: Color intensity of untreated and plasma-treated cotton fabrics

Optimization via Box Behnken Design and Verification of Optimized Values

The optimal conditions for obtaining the maximum dye uptake response were predicted using the Box-Behnken Design. A total of 17 responses were collected for the optimization stage, as shown in Table 6. This includes the 5 center points and the average response from the triplicates of the 12 combinations of parameters generated by the software.

Table 6: Experimental factors and responses for Box-Behnken design

	Factor 1	*Factor 2*	*Factor 3*	*Response*
Run	**Plasma Treatment Time (Seconds)**	**Oxygen Flow Rate (LPM)**	**Dye Bath Exposure (Minutes)**	**Dye Uptake Efficiency (%)**
1	30	3	30	6.69*
2	90	9	15	7.42*
3	30	15	30	6.75*
4	60	9	30	7.95
5	90	3	30	7.58*
6	60	9	30	8.61
7	90	15	30	7.81*
8	60	15	45	8.64*
9	60	9	30	8.05
10	30	9	15	5.80*
11	60	9	30	8.88
12	30	9	45	8.85*
13	90	9	45	10.18*
14	60	9	30	8.93
15	60	3	45	9.00*
16	60	3	15	6.47*
17	60	15	15	6.56*

* Represents the average response from the triplicate experimental runs

Several tests, including the sequential model sum of squares, lack of fit test, and model summary statistics, were performed to evaluate the model's fitness and adequacy as presented in Table 3.5. All tests suggested that the obtained data fits well to a quadratic model. For instance, the p-value of 0.0126 for the quadratic vs. 2FI is significant suggesting that the model fits well to a quadratic model. Similarly, the p-value of 0.6784 for the lack of fit test suggested that it fits well to a quadratic well as not significant value for lack of fit implies that there is no evidence to show that the model does not explain the variation in the responses. In addition, the model summary statistics presented that the quadratic model

was the best fit evident from its higher R^2 and adjusted R^2 values compared to other models.

Table 7: Results of sequential model sum of squares, lack of fit test, and model summary statistics

Test	Source	Sum of Squares	Df	Mean Square	F-value	p-value	
Sequential model sum of squares	Quadratic vs 2FI	3.94	3	1.31	7.74	0.0126	Suggested
Lack of fit	Quadratic	0.3442	3	0.1147	0.5430	0.6784	Suggested
		Std. Dev.	**R^2**	**Adjusted R^2**	**Predicted R^2**	**PRESS**	
Model summary statistics	Quadratic	0.4122	0.9454	0.8752	0.6866	6.83	Suggested

Model reduction was applied to remove insignificant factors and improve the model. The ANOVA results of the reduced model for responses are shown in Table 8. The model F-value of 28.33 and p-value of < 0.0001 indicate the significance of the established model. It is also evident from the results that while A, C, and B^2 are significant model terms, B is statistically insignificant. This implies that further increasing the oxygen flow rate may not result in a positive relationship with the dye uptake efficiency as it may impose oxidation damage on the sample. Moreover, the significant B^2 term suggests that the oxygen flow rate has a quadratic effect on the dye uptake efficiency, hence, the optimum value is situated along the curve rather than at the extremities. The lack of fit was also not significant which implies that there is no evidence to show that the model does not explain the variation in the responses.

Equation 3.1 shows the established model in terms of coded factors. The final equation, which incorporates the significant input variables determined after model reduction, can be used to make predictions about the response (dye uptake efficiency) for given levels of each factor (A. plasma treatment time, B. oxygen flow rate, C. dye bath exposure). Additionally, it is useful for identifying the relative impact of the factors by comparing the factor coefficients.

$$Dye\ Uptake\ Efficiency = 2.52979 + 0.020417A + 0.43B + 0.086833C - 0.023866B^2 \ldots\ldots (Eqn.\ 1)$$

Table 8: ANOVA results of the established model for responses

Source	Sum of Squares	Df	Mean Square	F-value	p-value	
Model	19.70	4	4.92	28.33	< 0.0001	significant
A-Plasma Treatment Time	3.00	1	3.00	17.26	0.0013	
B-Oxygen Flow rate	0.0000	1	0.0000	0.0003	0.9867	
C-Dye Bath Exposure	13.57	1	13.57	78.07	< 0.0001	
B^2	3.13	1	3.13	17.98	0.0011	
Residual	2.09	12	0.1739			
Lack of Fit	1.24	8	0.1551	0.7343	0.6718	Not significant
Pure Error	0.8451	4	0.2113			
Cor Total	21.79	16				

In the validation of statistical analysis, it is crucial that the data come from a normal distribution. One way to assess the normality of data is to have a normal distribution of residuals, which can be achieved if the residuals fall approximately along a straight line (Haji 2017). The normal probability plot of the residuals is presented in Figure 12, which shows that the residuals are along the linear graph.

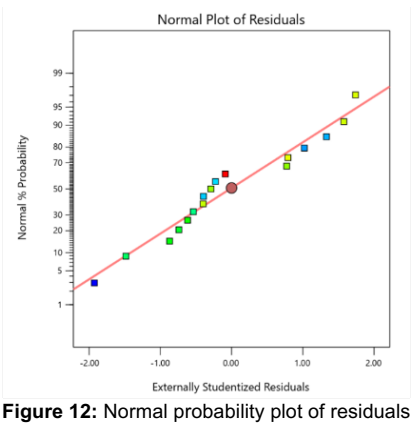


Figure 12: Normal probability plot of residuals

The surface plots, indicating the simultaneous effects of factors AB, AC, and BC on the dye uptake efficiency, are shown in Figure 13. Figures 13a and 13c revealed that increasing the oxygen flow rate led to an initial increase in the dye uptake efficiency. However, further elevating the oxygen flow rate resulted in a decline in efficiency. On the other hand, increasing efficiency for increasing plasma treatment time and dye bath exposure is observed for all surface plots.

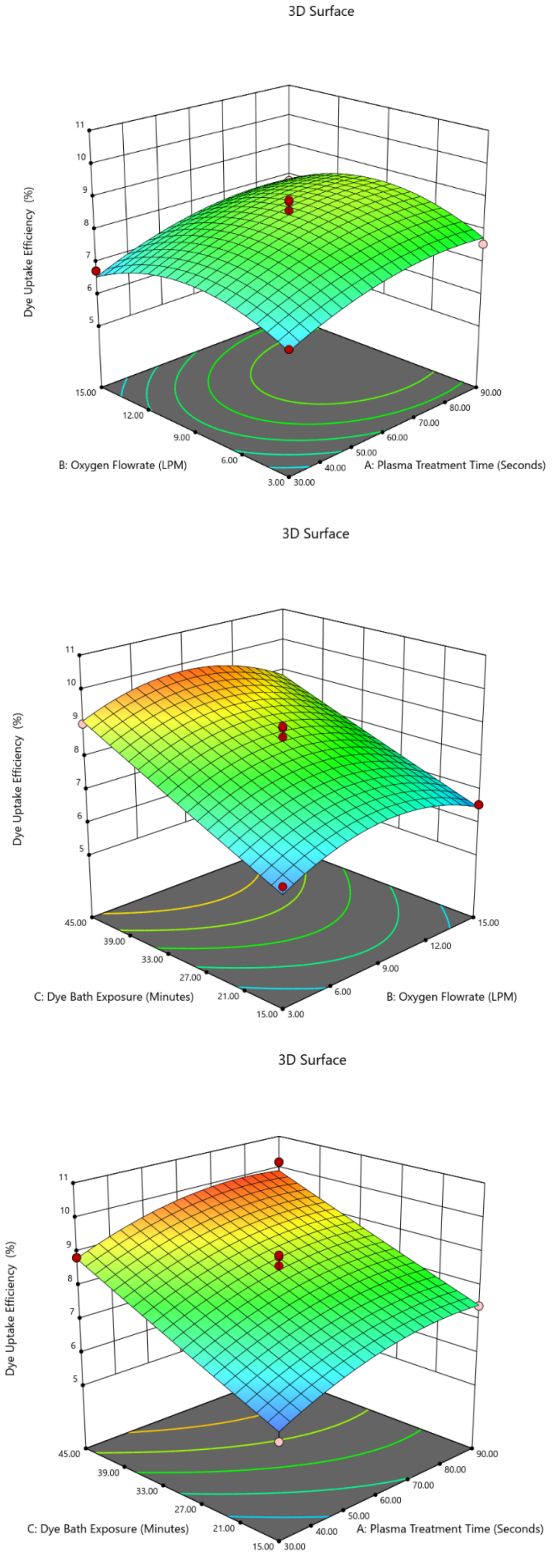


Figure 13: Surface plots of the effects of various interaction of independent variables. a) AB interaction b) AC interaction and c) BC interaction

To obtain an optimized predicted response, the levels of all experimental factors were set to be "in range", whereas the response was set to maximum. The software presented 10 various solutions; Table 9 displays the solution with the highest predicted dye uptake efficiency response, representing the optimal combination of experimental factor values for achieving the maximum response, which was chosen as the optimized parameters for this study.

Table 9: Software-predicted optimal conditions to obtain maximum dye uptake efficiency

Plasma Treatment Time (Seconds)	Oxygen Flow Rate **(LPM)**	Dye Bath Exposure **(Minutes)**	Dye Uptake Efficiency **(%)**	Desirability
90	9	45	10.21	1

Using the optimized parameters: 90 s plasma treatment time, 9 LPM oxygen flow rate, and 45 mins dye bath time, the predicted response was verified experimentally. As shown in Table 10, the average dye uptake response obtained after performing three experimental trials was 9.85%. The proximity of the predicted and experimental values, with a relative error of 3.56%, indicates that the RSM-derived model is suitable for accurately describing the relationship between the experimental factors and the dye uptake response.

Moreover, Figure 14 shows a comparison of the dye uptake efficiency of untreated cotton textiles and the plasma treated cotton textiles using the software- predicted optimal parameters. Results show that the dye uptake efficiency of raw cotton is 3.56%, signifying a 177% improvement under the optimized conditions.

Table 10: Experimental verification of the software-predicted value for optimum dye uptake efficiency

Trial 1 (%)	Trial 2 **(%)**	Trial 3 **(%)**	Mean **(%)**	Relative error **(%)**
9.89	10.03	9.63	9.85	3.56

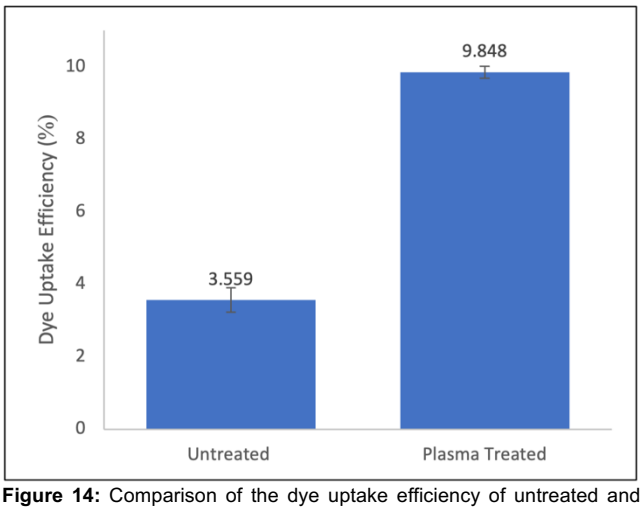


Figure 14: Comparison of the dye uptake efficiency of untreated and plasma treated cotton

CONCLUSION

A modified Atmospheric Pressure Plasma Jet (APPJ) system was used to generate Ar-O_2 plasma, confirmed by Optical Emission Spectroscopy to contain neutral and singly ionized argon and oxygen species. The measured electron and gas temperatures (8,444 K and 432 K) indicate non-thermal plasma.

SEM images showed surface etching on cotton textiles, likely due to the removal of wax and sizing agents, resulting in increased roughness and surface area. FTIR analysis revealed enhanced O-H, C-H, and C-O absorption peaks, indicating higher cellulose purity and increased hydrophilicity due to oxygen-containing polar groups.

As plasma treatment time increased, water contact angle, absorbance rate, and fabric weight decreased, while water and dye absorbency improved—indicating enhanced hydrophilicity and dyeability.

Plasma treatment time, oxygen flow rate, and dye bath exposure significantly affected dye uptake. RSM optimization identified ideal conditions (90 s, 9 LPM, 45 mins) yielding 10.21% dye uptake. Experimental results showed 9.85%, a 3.56% deviation from prediction, and a 177% improvement over untreated cotton (3.56%).

These results demonstrate the potential of APPJ treatment as a sustainable, energy-efficient, and cost-effective method for enhancing cotton textile processing.

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CONFLICT OF INTEREST

The author has no conflicts of interest to declare that are relevant to the content of this article.

CONTRIBUTIONS OF INDIVIDUAL AUTHORS

EJTP contributed to the conceptualization, methodology, formal analysis, investigation, original draft preparation and review and editing of this work. MMMR and GMM contributed to the methodology, as well as the review and editing of this work.

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