

## Optimisation of Nitrogen Plasma Exposure Time for Surface Modification of Cotton Fibre

Ainul Hafiza Abd Hair,<sup>a,b</sup> Kushairi Mohd Salleh,<sup>c,d,\*</sup> Nyak Syazwani Nyak Mazlan,<sup>a</sup> Mohamad Khalid Khairunnisa-Atiqah,<sup>a</sup> Noorain Purhanudin,<sup>e</sup> Anin Sofya Mohd Akhiri,<sup>f</sup> Sarani Zakaria,<sup>a</sup> and Rozidawati Awang<sup>f,\*</sup>

Surface modification *via* plasma treatment is useful in improving textile-based wound dressing functionality. This study was conducted to optimise the nitrogen plasma exposure time and its effect on the cotton surface (CS) properties at a constant nitrogen flow rate of 20 sccm for 5 to 30 min. The optimisation was done by analysing the alteration in morphology, functional group composition, crystallinity phase, electrokinetic potential, and colour of CS as subjected to nitrogen plasma. CS experienced an etching effect due to the presence of microcracks on its surface, with its electrokinetic potential becoming less negative, ranging from -5.51 to -8.05 mV. Then, the nitrogen functional group was detected on CS ranging from 2.9% to 4.5%, with its whiteness index reduced to 8.67% compared to the pristine cotton. As a result, 20 min was selected as the optimum exposure time for surface treatment. An exposure time of 30 min showed an early sign of degradation, which reduced its crystallinity index by 11.1%. Apparently, the CS is activated as exposed to the nitrogen plasma and experiences slight changes in its molecular structure without affecting its bulk properties.

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Contact information: a: Bioresources and Biorefinery Laboratory, Department of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia; b: Centre of Foundation Studies, Universiti Teknologi MARA, Cawangan Selangor, Kampus Dengkil, 43800 Dengkil, Selangor, Malaysia; c: Bioresource Technology Division, School of Industrial Technology, Universiti Sains Malaysia, Penang 11800, Malaysia; d: Green Biopolymer, Coatings & Packaging Cluster, School of Industrial Technology, Universiti Sains Malaysia, Penang 11800, Malaysia; e: Department of Earth Sciences and Environment, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia; f: Department of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia;

\* Corresponding author: kmsalleh@usm.my; rozida@ukm.edu.my

### INTRODUCTION

Cotton gauze is recognised as the safest and most acceptable biopolymer textile for medical applications due to its ubiquity, low cost, biocompatibility, and longstanding touch with humans, especially in the wound management system. Its affinity towards skin with excellent tensile strength and high hygroscopic capability can ideally be employed as a wound dressing, as it can remarkably absorb wound exudate and protect an open injury from further complications. By far, cotton gauze has become one of the most effective textile-based wound dressings that exploit the intrinsic properties of cellulose fibres (Abbasipour *et al.* 2014). Despite its usefulness, cotton gauze is considered a “dry” wound dressing, as it provides minimal or no therapeutic effect on wound healing. Thus, the wound dressing properties of cotton gauze should be modified or upgraded to perform

further than its primary function. Furthermore, ensuring that the modification will not degrade the existing properties of cotton gauze is of utmost importance. Consequently, this will ensure that the cotton gauze will continue to function as expected despite the changes that have been made.

For this reason, surface modification *via* non-thermal plasma treatment can be employed to modify cotton gauze without suffering total changes in its bulk properties (Kan and Man 2018; Tudoran *et al.* 2020). Plasma is created once any particular gas is subjected to high heat or a strong electromagnetic field (Thi *et al.* 2020). When exposed to high energy forces, these gases are compelled to be partly ionised and possess high electrical conductive ability composed of free ions, electrons, photons, and other reactive elements (Tudoran *et al.* 2020). The highly reactive radicals generate molecules in plasma that will aggressively radiate, collide, and bombard the surface, altering roughly a few nanometres depth of the treated cotton surface (Rumi *et al.* 2022). Furthermore, as a dry process, plasma treatment should be more environmentally friendly because it does not use hazardous chemicals or generate dangerous by-products after treatment (Gao *et al.* 2019; Ji *et al.* 2020). Hence, plasma may become one of the options for switching or reducing human reliance on non-environmentally friendly wet chemical treatment for surface modification (Zhao *et al.* 2021). The impact of plasma treatment on material surface relies on the type of gases, exposure period, and energy source utilised to create plasma (Ayesh *et al.* 2022).

Regarding improving the medicinal value of cotton gauze as a wound dressing, nitrogen plasma treatment has been shown to be suitable for altering heat-sensitive materials such as cotton due to its non-thermal and low-pressure properties, as well as maintaining its bulk properties (Kert *et al.* 2021). Nitrogen plasma can be exploited as a pre-treatment step in which plasma-treated cotton can become a substrate for the subsequent post-modification process, expanding its medical functionality as a wound dressing. Therefore, plasma-treated cotton more readily undergoes the post-modification process, in which its altered surface can be coated, soaked, or impregnated with therapeutic biomaterials, including drugs, nanoparticles, plant extract, and polymers *via* chemical or physical post-modification (Eswaramoorthy and McKenzie 2017). In most cases, nitrogen plasma has been found to introduce a new functional group (-NH) and alter the existing functional groups (hydroxyl, carboxyl) due to surface ablation (Prysiashnyi *et al.* 2013). As a result, these functional groups become the anchoring sites for the attachment of therapeutic materials (Shahidi *et al.* 2010) such as metal oxide, nanoparticles, drugs, or polymers on the cotton surface, thus producing cotton gauze that benefits wound treatment.

In addition, nitrogen plasma can also lead to surface ablation and create a dry etching effect (Pransilp *et al.* 2016), as indicated by microcracks, slits, and grooves on the treated surface, hence changing the microstructure surface while retaining the bulk properties of the treated material. The etched surface results due to the collision of free radicals and ion bombardment during plasma treatment (Shahidi and Ghoranneviss 2016), thus increasing the surface area of the treated material (Eswaramoorthy and McKenzie 2017). The microcracks and large surface area on the treated cotton surface provide a better space for the mechanical interlocking between biomaterials (nanoparticles, drugs, polymers) and cotton surface (Zhang *et al.* 2016). Moreover, a large surface area is also essential to help increase the number of biomaterials attached to the crack's gaps on the treated surface, significantly enhancing the medicinal properties of the treated cotton. In addition, it has also been demonstrated that nitrogen plasma can improve wettability, adhesion, and surface charge, making cotton gauze ready as a wound dressing while

retaining its intrinsic properties (Vinisha Rani *et al.* 2018). Furthermore, plasma treatment of cotton is also effective for decontamination, sterilisation (Liu *et al.* 2021), cross-linking (Zhang *et al.* 2022), and surface cleaning purposes (Kramar *et al.* 2018).

Although the use of plasma treatment for the surface modification of cotton gauze is not new, only a few works have described the potential of nitrogen plasma as a pre-treatment tool in transforming cotton gauze into a wound dressing with healing properties (Przyshchyni *et al.* 2013; Shahidi *et al.* 2014; Zhou *et al.* 2017). Therefore, in this work, the surface of cotton gauze was treated using radio-frequency plasma-enhanced chemical vapour deposition-nitrogen plasma (RF-PECVD-NP), and the changes in its surface morphology, chemical composition, crystallinity index (CI), surface charge, and colour were evaluated. The chosen analysis was done to study the optimisation of nitrogen plasma exposure time in modifying the surface of cotton gauze. This approach will further benefit subsequent post-treatment modification of the cotton gauze as a wound dressing with healing properties. The obtained data can be used as a reference to assist future post-treatment concerning the accessibility and biocompatibility of cotton in combination with other biomaterials, thus upgrading the cotton properties.

## EXPERIMENTAL

### Materials

The woven cotton gauze (10 cm × 10 cm, 24 ply) with 20 × 24 mesh count was purchased from MyMedic Innovation Sdn. Bhd., Malaysia. After plasma treatment, the samples were kept in an air-tight container, and the treated surfaces were marked to specify the surface that went for an analysis.

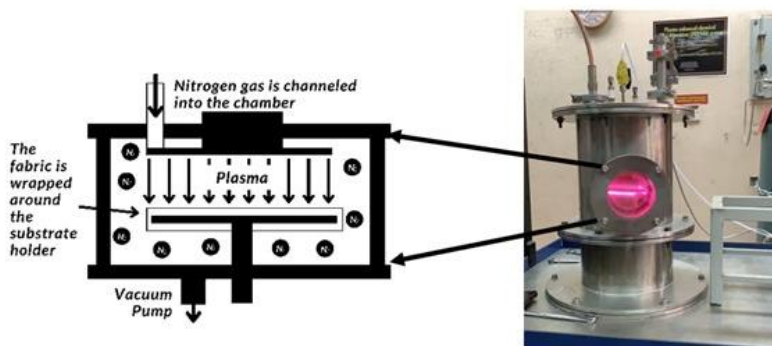


Fig. 1. Schematic diagram of RF- PECVD-NP system

### RF-PECVD-NP Treatment Setup

The cotton gauze was subjected to nitrogen plasma treatment produced by the RF-PECVD-NP system at 5 min (G5), 10 min (G10), 15 min (G15), 20 min (G20), and 30 min (G30) with a fixed nitrogen gas (99% purity) while maintaining a fixed nitrogen gas flow rate of 20 sccm and a radio frequency (RF) power output of 50 W. The schematic diagram depicted in Fig. 1 illustrates the configuration of the in-house RF-PECVD-NP system, which facilitated the generation of pink-colored nitrogen plasma. Prior to plasma treatment, the cotton gauze was centrally positioned within the chamber on the substrate holder, situated between two electrodes, at an electrode gap of 2.0 cm indicating the distance between the substrate holder and the shower head gas emitter. The chamber with a volume of 13,090 cm<sup>3</sup>, was initially evacuated using a Pascal Series Adixen rotary pump until it

reached a base pressure of  $3.4 \times 10^{-3}$  mbar, and continuously pumped with turbo Edward pump until  $3.0 \times 10^{-5}$  mbar to ensure tiny particles and dirt were completely removed from the chamber. Subsequently, the working pressure chamber for the nitrogen plasma treatment was set to 0.6 mbar at room temperature, operating at frequency of 13.56 MHz.

### Field Emission Scanning Electron Microscopy-Energy-Dispersive X-Ray Spectroscopy Mapping

The surface morphological properties of the untreated cotton gauze (UCG) and treated cotton gauze (TCG) exposed to the RF-PECVD-NP treatment at 5 min (G5), 10 min (G10), 15 min (G15), 20 min (G20), and 30 min (G30) were observed using a scanning electron microscope (Zeiss Supra 55VP). Before the treatment, all samples were sputter-coated with gold before scanning and the magnification used in this study was 20000 x with the beam energy of 15 KeV (Thi *et al.* 2020). Besides field emission scanning electron microscopy (FESEM), the elemental information and purity of the UCG4 and TCG at different exposure times were also investigated using energy-dispersive X-ray spectroscopy (EDX).

### Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy

The UCG and TCG samples at different exposure times (5, 10, 15, 20, and 30 min) were analyzed using attenuated total reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy (Bruker, Massachusetts, United States). The IR spectrum of each sample was recorded at an average of 64 scans with  $2 \text{ cm}^{-1}$  resolution ranging between 4,000 and 500 cm.

### X-Ray Diffraction

The X-ray diffraction (XRD) characterisation of the crystalline nature and phase changes on the UCG and TCG samples was performed using a diffractometer (Bruker AXS D8 Advance, Germany). The amount of sample was filled in the sample holder and then scanned with a diffraction angle ( $2\theta$ ) from  $5^\circ$  to  $60^\circ$ , Cu-K $\alpha$  radiation ( $\lambda = 0.15418 \text{ nm}$ ), step size of 0.04 time per step at 1.20 s (fast detector), a 1-D fast detector, and scan type of locked couple ( $\theta$  to  $2\theta$  scan).

### Zeta Potential Analysis

The surface charge properties of the UCG and TCG samples were measured using a Zetasizer analyser (Malvern Instruments). The UCG and TCG samples (0.2% (w/w)) were vibrated for 48 h in an equilibrated cell, and the zeta potential analysis was performed at pH 6.8 and  $25^\circ\text{C}$ .

### Colouration Measurement

The effect of the RF-PECVD-NP treatment on colour changes of UCG and TCG at different plasma exposure times was evaluated using a benchtop spectrophotometer (Agera, Hunter Lab). The colour variation was analysed using the  $L^*$ ,  $a^*$ , and  $b^*$  parameters, in which  $L^*$  is for lightness,  $a^*$  is for red or green,  $b^*$  is for yellow or blue, and  $dL^*$  values (Haji *et al.* 2021). The measurements were performed on a white background, where the  $L^*$  (86.46),  $a^*$  (-0.19), and  $b^*$  (-0.32) values of UCG (G0) were set as the standard background parameters. According to the Commission Internationale de l'Eclairage (CIE),  $L^*$  is a measurement for lightness (- = darker, + = lighter),  $a^*$  is for the red and green coordinates (+ = redder, - = greener), and  $b^*$  is for the yellow and blue

coordinates (+ = yellower, - = bluer). Therefore, the measured values for  $L^*$ ,  $a^*$ , and  $b^*$  can be positive (+) or negative (-), depending on which coordinate that respective colour is more visible after the treatment. As referred to in the CIE ( $L^*$ ,  $a^*$ , and  $b^*$ ) colour scheme (Ahmed *et al.* 2020), the values for each coordinate are as follows:  $L^*$  values range from 0 to 100, and both  $a^*$  and  $b^*$  values range from -128 to +127. The obtained values from  $L^*$ ,  $a^*$ , and  $b^*$  were used to calculate the whiteness index (WI) of all samples by Eq. 1

$$WI = 1\sqrt{(100 - L)^2 + a^2 + b^2} \quad (1)$$

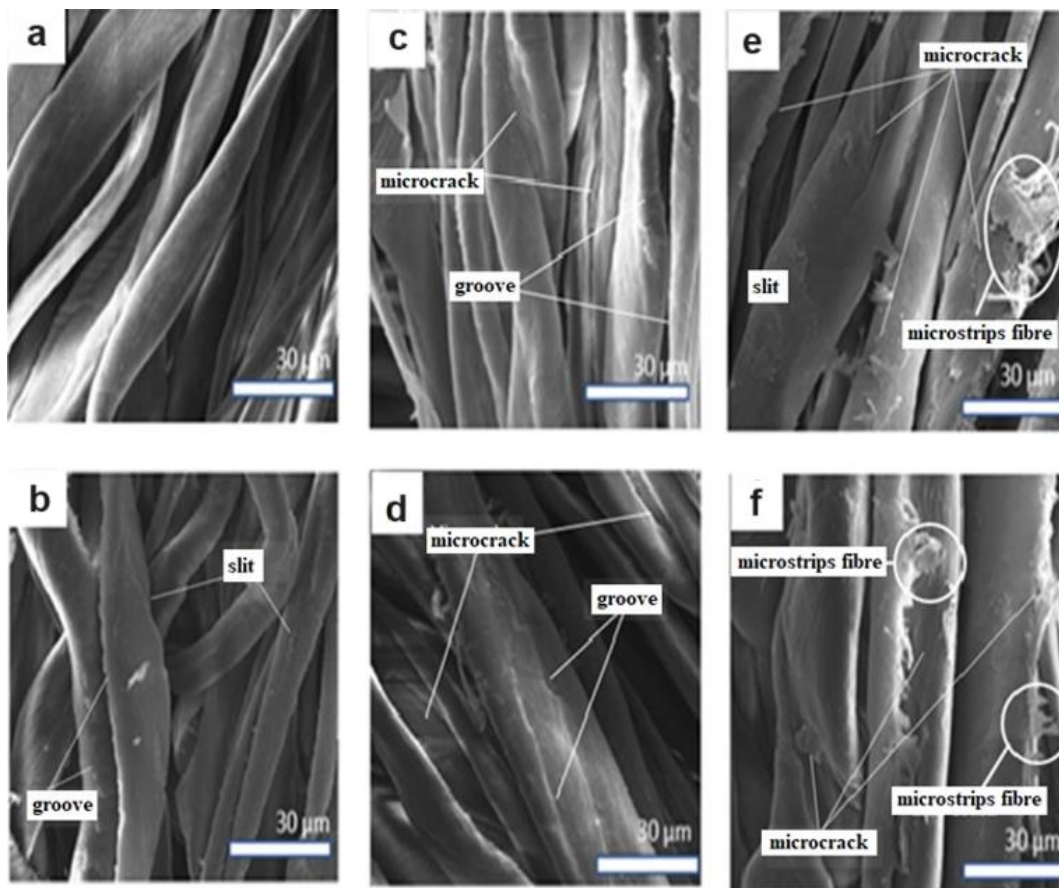
## RESULTS AND DISCUSSION

### Surface Morphological and Chemical Properties

FESEM and EDX are among the most reliable methods that can be used to study the effect of plasma treatment on the morphological and elemental changes in the plasma-treated cotton, respectively. The FESEM images in Fig. 2 (a–e) show the surface morphology of TCG at different exposure times. Morphologically, the surface of UCG (G0) was smooth with a tidy and neat striation feature composed of a mutual intertwined bundle of cellulose fibres (Fig. 2a). After plasma treatment, the TCG exposed from 5 to 30 min (Fig. 2b–f) exhibited a slit, groove, and microcrack appearance with different intensities on its surface due to the surface ablation and an etching effect introduced by the nitrogen plasma treatment (Przyaszny *et al.* 2013; Kert *et al.* 2021; Hafiza *et al.* 2023). The etching effect can be observed as early as at 5 min (G5) of exposure time (Fig. 2b). In Fig. 2b, there was an occurrence of slits and grooves without any sign of microcracks on its surface. Then, at 10 min of exposure time (Fig. 2c), the treated surface started to experience microcracks with more slits and grooves appearing on its surface. Afterwards, these minor cracks, tiny grooves, and slits became more evident as the TCG was subjected to a 15-min (G15) exposure time (Fig. 2d).

Later, the TCG cotton surface morphology for G20 and G30 experienced more slits and grooves than G5, G10, and G15, as can be observed in Fig. 2e and Fig. 2f, respectively. The etching effect became intense and severe due to excessive surface ablation, which could be seen on the cotton surface as it was exposed to a more prolonged period of plasma treatment. The observed surface morphological changes on TCG can be explained, as a continuous exposure of nitrogen plasma to the surface of cotton gauze will allow more time for nitrogen plasma-generated reactive molecules (*e.g.*, nitrogen ions, electrons, photons, and other free radicals) to bombard the cotton surface, thus providing more noticeable etching and sputtering effects. Therefore, based on the FESEM images of TGC, plasma exposure time can significantly affect the severity of etched surfaces. The persistent cotton exposure towards plasma treatment will cause a higher degree of etching effect and can even reduce its mechanical properties (Kert *et al.* 2021; Klébert *et al.* 2021). In addition, when the exposure time increased from 15 to 20 min (Fig. 2e), the etching effect on the treated cotton became excessive, with the appearance of microfibril cotton strips on the treated cotton. Later, the etching effect became more prominent as the grooves, microcracks, and microfibril cotton strip formation became more severe for G30 (Fig. 2f). The microfibril cotton strips could most likely be stripped away or torn off from the cotton surface, indicating the early stage of polymer degradation due to the severe etching effect following prolonged exposure to plasma treatment (Klébert *et al.* 2021).





**Fig. 2.** FESEM images of (a) UCG (G0) and TCG at (b) 5 min (G5), (c) 10 min (G10), (d) 15 min (G15), (e) 20 min (G20), and (f) 30 min (G30)

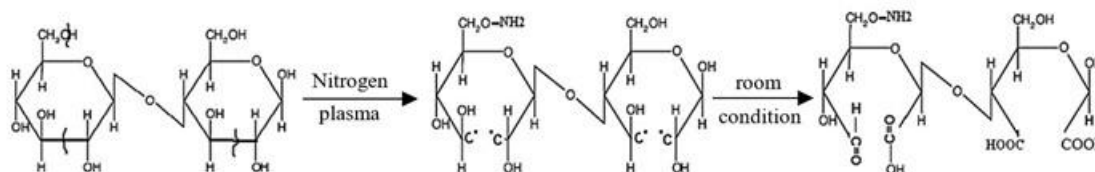
However, besides an early sign of degradation, the observation of microfibril cotton strips in the FESEM images indicates that plasma treatment can also cause fibre fibrillation by slicing the fibre surface into smaller fibres or microfibrils. Additionally, fibres such as cotton are more prone to be fibrillated due to their chemical composition and intrinsic layered surface morphology. In this study, the samples treated with plasma for up to 20 min showed an early sign of surface fibrillation, and the changes became more evident as the exposure time increased up to 30 min with fixed plasma power and nitrogen amount. This surface ablation occurs when the high-energy reactive species generated in plasma interact with the cotton fibre surface upon surface modification. According to Ayesh *et al.* (2022), the fibrillation process by plasma treatment exposed the microfibrillar surface texture of the secondary wall of the treated surface as its primary wall had been etched away during plasma treatment.

As the FESEM images detected and validated the etching effect, the EDX mapping analysis was further done to study how nitrogen plasma can influence the elemental composition changes in plasma-treated cotton gauze. Table 1 presents complete quantitative changes in the elemental composition, mainly on carbon and oxygen elements on UCG (G0) with newly introduced nitrogen element in TCG at 5 min (G5), 10 min (G10), 15 min (G15), 20 min (G20), and 30 min (G30). Even though cotton is made up of carbon, oxygen, and hydrogen, only the peaks for carbon and oxygen were identified, as shown in Table 1, due to the inability of EDX analysis to detect the hydrogen atom.

**Table 1.** Carbon, Oxygen, and Nitrogen Content in UCG and TCG Samples

Sample	Carbon (%)	Oxygen (%)	Nitrogen (%)
G0	56.4	43.6	NONE
G5	55.3	42.2	2.5
G10	54.4	42.4	3.2
G15	51.9	43.8	4.3
G20	50.1	44.7	5.2
G30	49.8	47.6	2.6

According to the results in Table 1, the UCG of G0 was observed with the highest carbon content of 56.4% and the lowest oxygen content of 43.6%, which are comparable to a previous study (Thi *et al.* 2020). This observation is in agreement with Ayesht *et al.* (2022) and Thi *et al.* (2020), whose EDX studies reported the presence of nitrogen on the plasma-treated cotton fabric after plasma treatment. During plasma treatment, electrons and ions are accelerated to the exposed cotton, thus increasing the reactivity of the plasma-treated surface. Then, once the TCG is exposed to open air, the response of the reactive treated surface with the free radicals in the atmosphere leads to surface activation (Shahidi *et al.* 2014, 2018), as elucidated in Fig. 3. After plasma treatment, the carbon content of TCG appeared to decrease gradually, while the oxygen content oppositely increased over exposure time due to surface oxidation. The decrease in carbon content, an increase in oxygen percentage, and the presence of nitrogen could be attributed to the presence of high-intensity numerous oxygen and nitrogen functional groups, such as  $-O-C=O$ ,  $-O-O-$ ,  $C=O$ ,  $C-O$ ,  $-COOH$ ,  $-COH$ , and  $-NH$  on the surface of TCG (Kramar *et al.* 2014).

**Fig. 3.** Surface of TCG at ambient temperature after plasma treatment

According to Pranslip *et al.* (2016), Zhou and Kan (2014), and Ibrahim *et al.* (2017a), nitrogen plasma can decrease or increase the cotton's existing elements (C and O) due to surface oxidation and introduce nitrogen elements on the treated cotton. These changes lead to more nitrogen- and oxygen-based functional groups on the treated cotton, which significantly induce chemical or physical bonding between bioactive molecules (drugs, polymers, and nanoparticles) and the surface of cotton gauze, producing a multifunctional wound dressing that benefits wound healing. Therefore, the observed differences in the elemental composition of TCG are most likely due to the reaction of plasma reactive molecules with the cotton surface, which can significantly intensify and/or introduce various functional groups on the surface of cotton gauze. This matter is further discussed in the FTIR analysis.

### Surface Chemical Composition

The FTIR analysis was performed to compare the differences in surface chemical functionality between UCG and TCG at different exposure times. Their differences in surface chemistry are shown in Fig. 4, in which the FTIR spectra of the UCG and TCG samples seem to be similar, except for the changes in their intensities. The UCG displays several transmittance regions that correspond to the typical cellulose characteristic: C-H

stretching at 2,800 to 3,000  $\text{cm}^{-1}$ , O-H stretching at 3,300 to 3,400  $\text{cm}^{-1}$ , C-O-C at 1,050 to 1,060  $\text{cm}^{-1}$ , and C-O at 1,080 to 1,300  $\text{cm}^{-1}$ , which display similar results with the study on native cellulose reported by Hospodarova *et al.* (2018). In addition, the peaks observed in the 890 to 895  $\text{cm}^{-1}$  range are related to cellulosic-glycosidic linkages that indicate the bonding between cellulose monomers in the native cellulose (Fazeli *et al.* 2019). After plasma treatment, the spectra for all TCG samples were identical to UCG (G0). Nonetheless, some regions, specifically at 700 to 1,100  $\text{cm}^{-1}$  (C-H stretching), 1,200 to 1,400  $\text{cm}^{-1}$  (C-O stretching), 2,800 to 3,000  $\text{cm}^{-1}$  (O-H stretching overlapped with N-H bending), and 3,200 to 3,500  $\text{cm}^{-1}$  (O-H stretching) were intensified with the increase of plasma exposure time.

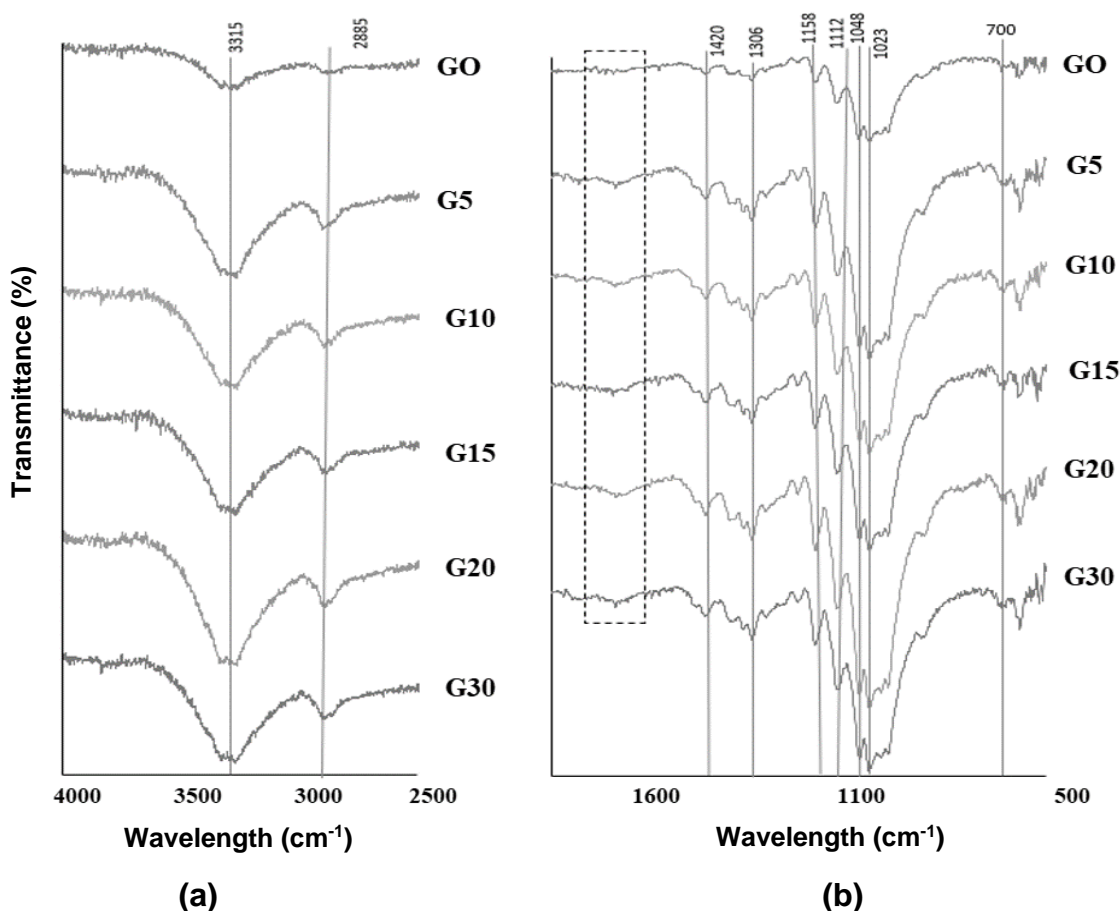
The transmittance intensity of TCG started to increase as early as 5 min (G5) and became further intense as the cotton gauze was subjected to plasma treatment from 10 min (G10) to 20 min (G20). Aboltakhty *et al.* (2018), Shahidi *et al.* (2018), and Shahidi *et al.* (2014) reported that the 2,800 to 3,400  $\text{cm}^{-1}$  region intensified and became more expansive due to the introduction of N-H stretching that overlapped with O-H stretching (Ibrahim *et al.* 2017). According to Shahidi *et al.* (2018), the reaction between the active species induced by the plasma in the gas phase and the fabric surface atoms generated these functional groups on the cotton surface. Then, some of the observed peaks, including 1,023, 1,048, 1,112, 1,158, 1,308, 1,420, and 1,650  $\text{cm}^{-1}$  that intensified in all TCG samples corresponded to C-O stretching, C-N stretching, CO-O-CO stretching, C-O stretching, C-H bending, O-H bending, and N-H bending that overlapped with C=N stretching, respectively. Moreover, a small narrow transmittance band at 1,650  $\text{cm}^{-1}$  reveals the existence of an amide group, which confirms that nitrogen plasma treatment adds an amide group to the TCG surface (Zhou and Kan 2014). These observations are due to the collision of the cotton with the ion bombardment resulting from the active species, such as  $\text{N}_2^+$ ,  $\text{N}_2$  (excited), N, and  $\text{N}^+$  particles, as well as the free electron in nitrogen plasma. This can increase or introduce new functional groups, such as N-H (during plasma treatment), while intensifying the oxygen-based functional group on its surface due to oxidation when exposed to the air.

The intensity of the functional group on the treated cotton sample at 30 min (G30) started to exhibit a slight decrement in its transmittance intensity compared with other treated samples. This is because the cotton surface has reached its saturation point at 20 min of plasma exposure time. Therefore, plasma exposure after 20 min will decrease the functional group intensity. No more functional groups can be introduced or added to its surface due to the severe surface etching during plasma treatment, consequently suffering early signs of degradation. Afterwards, the observed peaks from 700 to 800  $\text{cm}^{-1}$  and 900 to 1,200  $\text{cm}^{-1}$  in all plasma-treated cotton samples confirm that plasma treatment can maintain the  $\beta$ -glycosidic linkage ( $\text{C}_1\text{-O-C}_4$ ) and the cyclic ether linkage ( $\text{C}_3\text{-O-C}_1$ ) in its cellulose chain, respectively (Fazeli *et al.* 2019). These empirical findings are consistent with previous literature, as both linkages are crucial and among the most essential typical chemical interactions in the cellulose backbone (Caschera *et al.* 2014; Fazeli *et al.* 2019). This observation indicates that plasma treatment only permits the initiation of surface chemical reactions on cotton while maintaining its typical backbone chemical interaction. Hence, the TCG clarifies that plasma treatment affects only the uppermost atomic layers of the cellulose surface, and most of its intrinsic molecular structure remains unaffected (Zhou *et al.* 2016).

Based on the EDX and FTIR analysis, the bombardment of plasma reactive molecules on the cotton had modified the cellulose surface by changing its surface element,



leading to various functional groups including -OH, -COOH, -CH, and -NH appearing on its surface. As a result, the cotton surface has more reactive sites than UCG, increasing its affinity towards preferred biomaterials upon instilling them on its surface, as reported by Shahidi *et al.* (2018), Vinisha Rani *et al.* (2018), and Honarvar *et al.* (2017). In these studies, plasma treatment as a pre-treatment method assisted the adhesion and attachment of zinc oxide (Shahidi *et al.* 2018), graphene oxide (Vinisha Rani *et al.* 2018), and essential oil (Honarvar *et al.* 2017) on the treated cellulose, thus improving the medicinal properties of the treated cellulose that benefited in wound treatment. In addition, Vosmanská *et al.* (2015) reported that the presence of a new reactive site due to plasma treatment had positively affected both impregnation of chitosan and silver chloride on the plasma-treated cotton that exhibited the growth skin pathogens, including *Escherichia coli* and *Staphylococcus aureus*.



**Fig. 4.** FTIR spectra of UCG (G0) and TCG at a) 4,000–2,500 cm<sup>-1</sup> and b) 1,600–600 cm<sup>-1</sup> for 5 min (G5), 10 min (G10), 15 min (G15), 20 min (G20), and 30 min (G30)

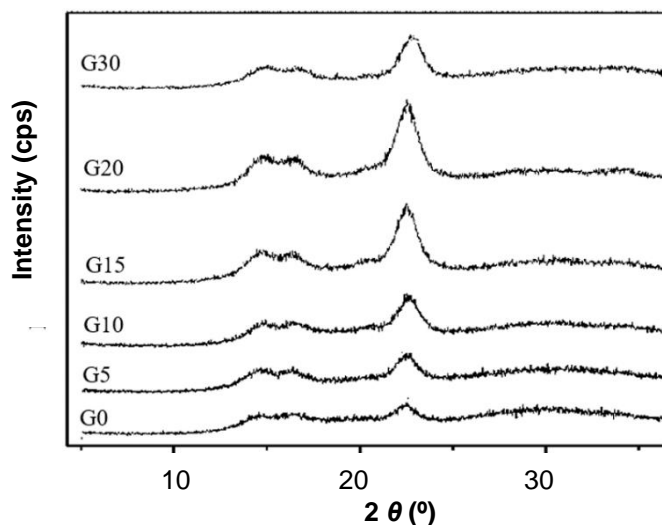
### Crystallinity Nature

The XRD analysis was used to observe the crystalline structure changes between UCG and TCG samples at different plasma treatment exposure times. The observed XRD result is displayed in Fig. 7 with a double-headed peak around  $2\theta = 16^\circ$ , corresponding to (110) and (110) crystallographic planes commonly associated with high cellulose content materials, such as cotton (Rumi *et al.* 2022). Then, a higher peak corresponding to the (002)

crystallographic plane of cellulose can also be detected at  $2\theta = 22.6^\circ$ . Compared to UCG (G0), both peaks at  $16^\circ$  and  $22.6^\circ$  for TCG intensified as the plasma treatment exposure time lengthened from 5 to 20 min and decreased when it was at 30 min with a slightly shifted of peak  $22.6^\circ$  towards a higher angle side. The obtained diffractogram was further used to calculate the percentage of CI for all samples, and the values are presented in Table 2. Similar to the increasing diffractogram pattern of Fig. 5, the CI for TCG also increased gradually as the sample was subjected to a continued plasma exposure time from 5 to 20 min and reduced at 30 min of exposure time.

**Table 2.** Crystallinity Index of UCG (G0) and TCG at 5 min (G5), 10 min (G10), 15 min (G15), 20 min (G20), and 30 min (G30)

Sample	CI
G0	35.96
G5	36.08
G10	37.79
G15	41.41
G20	43.34
G30	38.54



**Fig. 5.** XRD diffractogram for UCG and TCG at 5 min (G5), 10 min (G10), 15 min (G15), 20 min (G20), 30 min (G30)

The increment of the CI of TCG from 5 to 20 min treatment can be attributed to the removal of the amorphous region on the cotton surface due to the constant bombardment of plasma ions and reactive molecules generated from plasma (Nawalakhe *et al.* 2015). In this study, the removal of the cotton's amorphous region started when the etching effect occurred as early as 5 min (G5) of the nitrogen plasma treatment, leading to the loosening arrangement in the cellulose structure (Bhat *et al.* 2011) with its CI of 36.08%. Then, as the etching effect became more severe with the increase in exposure time, more amorphous regions were removed, thus increasing the crystallinity of the treated surface.

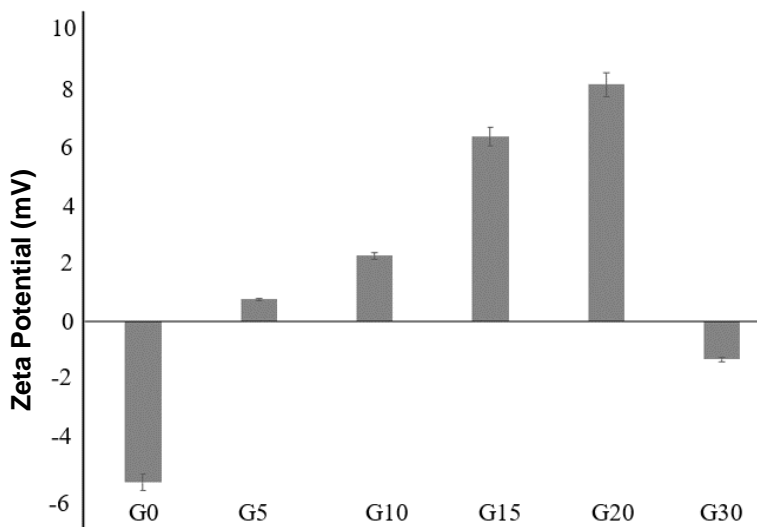
Next, at 30 min (G30), the intensity of the double-headed peak around  $2\theta = 16^\circ$  and peak  $22.6^\circ$  were decreased and became narrower due to further bombardment of nitrogen plasma continued destroying the amorphous region and, at the same time, started disrupting the crystalline area of cellulose, thus resulting in the lowest CI of 38.54%. Then, both peaks (double-headed peak around  $2\theta = 16^\circ$  and peak  $22.6^\circ$ ) were narrower and slightly shifted towards a higher angle in G30, which was related to the contraction and shrinking of the lattice parameter in its unit cell upon surface modification by plasma treatment (Saleem *et al.* 2021). According to Prabhu *et al.* (2017), this is due to the weakening of the intra and inter-molecular hydrogen bond which may eventually break thus disturbing the arrangement of the cellulose chain. This observation also agrees with da Silva *et al.* (2020), in which an etching effect due to the plasma treatment was claimed to change the cellulose surface at the molecular level, causing a weakly altered crystalline structure of the most superficially exposed cellulose.

In comparison to the amorphous region, the crystalline region in cotton is more resistant to being disrupted by the reactive molecules generated in the plasma because its arrangement is denser and more complex than the amorphous region. Nevertheless, prolonged plasma exposure led to a change in the geometry of the crystalline domain and a decrease in CI (Rumi *et al.* 2022), which caused degradation of the TCG at a 30-minute reaction time. Herein, the reduction of the CI of TCG can reveal the early stage of cellulose degradation, leading to the breakdown of cellulose structure.

In this regard, it is essential to choose the optimum plasma exposure time for surface modification so that cotton gauze will not suffer excessive structural degradation even though its surface has been bombarded with high-energy reactive molecules from plasma. Therefore, by referring to the CI in Table 2, 20-min exposure time with its CI was selected as the optimum exposure time, as the G30 sample showed a sign of degradation by having the lowest CI. This is also supported by the severity of the etching effect in the SEM image of the 30-min treatment (Fig. 2f) and the decrement of FTIR intensity, as seen in Fig. 4. In the end, the high appearance of cracks, slits, and grooves on the cotton surface resulting from plasma treatment disrupted the molecules' intermolecular arrangements, consequently affecting the CI of the G30 sample.

## Surface Charge

Surface treatment is typically performed to tune the functionality and characteristics of a solid surface while maintaining its bulk properties. According to Kramar *et al.* (2018), surface modification caused by plasma treatment on cotton surfaces can affect their electrokinetic properties (*i.e.*, zeta potential). This is because surface charge is directly related to the type and accessibility of the functional groups present on the cotton surface. In this regard, zeta potential analysis has become one of the most suitable methods for monitoring solid surface charge changes after experiencing a surface modification process *via* plasma treatment.



**Fig. 6.** Zeta potential of UCG (G0) and TCG at 5 min (G5), 10 min (G10), 15 min (G15), 20 min (G20), 30 min (G30)

The zeta potential measurement for all samples is displayed in Fig. 6. As shown, the UGC (G0) exhibited the lowest zeta potential with a negative value (-5.51 mV), thus indicating its negative surface charge. The UGC has a negative zeta potential because cellulose or pure cotton always has a high amount of carboxyl groups in its chemical structure (Aboltakhty *et al.* 2018; Kramar *et al.* 2018). After plasma treatment, the zeta potential of TCG started to become less negative or near to the positive value of 0.73 mV for G5, 2.23 mV for G10, 6.26 mV for G15, and 8.05 mV for G20 and declined to -1.32 mV for G30. Therefore, this study shows that nitrogen plasma treatment can induce changes in the electrokinetic properties of the treated surface. The zeta potential will become less negative for a longer plasma treatment exposure time due to the increasing amount of nitrogen-based functional groups on its surface, as determined in the FTIR analysis (Fig. 4). This observation is in agreement with Pransilp *et al.* (2016), in which the less negative zeta potential is encouraged by the fact that nitrogen plasma can introduce the formation of nitrogenous species-based functional groups on the treated surface. The higher the amount of nitrogen-based and other functional groups in the respective sample, the more cationic it will become (Aboltakhty *et al.* 2018), resulting in a positive zeta potential.

However, at 30 min (G30), the surface charge started to decrease or return to a negative value as its surface began to degrade due to the severe etching effect, as described in the previous sections. The severity for G30 is because more cotton surfaces are etched away by the severe bombardment of reactive molecules during plasma treatment, resulting in a sample with fewer functional groups and reduced surface charge. In addition, according to Aboltakhty *et al.* (2018) and Kramar *et al.* (2018), the severe etching effect can also eliminate the existing nitrogen-based functional groups on the cotton surface, hence reducing its surface charge and implying the negative zeta potential at 30 min (G30). Therefore, even though the respective cotton was subjected to the longest exposure time of 30 min, its surface charge could only display negative surface charges, resulting in a slightly lower zeta potential compared to the samples treated at 20 min and less.



## Color Measurement

Regarding colour, white cotton gauze is typically associated with purity, freshness, and cleanliness. Hence, its white colour is expected to be maintained even after plasma treatment. However, in this study, the pure white colour of cotton gauze changed into a white-yellowish tone, meaning that plasma treatment not only altered the cotton at its molecular surface but also changed the outer appearance of the cotton gauze. Even though the colour parameter does not directly contribute to the fabrication of cotton gauze as a functional wound dressing, one should expect that the appearance of plasma-treated cotton will not be as white as before plasma treatment due to surface modification. Therefore, colour measurement was done to study how the surface morphology and surface chemistry alteration by nitrogen plasma treatment on cotton gauze influence its whiteness index (WI). Table 3 displays the values for  $L^*$ ,  $a^*$  (red/green), and  $b^*$  (yellow/blue) parameters of colour, as well as the WI for both untreated (UCG) and treated cotton (TCG) at different exposure times.

**Table 3.** The Values of  $L^*$ ,  $a^*$ ,  $b^*$ ,  $dL^*$ , and WI for UCG (G0) and TCG at 5 min (G5), 10 min (G10), 15 min (G15), 20 min (G20), and 30 min (G30)

Sample	$L^*$	$a^*$	$b^*$	$dL^*$	WI
G0	87.52	-0.19	-0.32	87.52	87.51
G5	87.45	1.08	5.78	-0.99	86.41
G10	86.46	1.57	7.04	-1.06	84.66
G15	85.42	2.11	8.93	-1.04	82.77
G20	85.94	1.91	8.28	-0.52	83.57
G30	84.19	3.16	11.96	-2.26	79.93

Note: For sample G0,  $L^*$  is shown instead of  $dL^*$ .

After plasma treatment, the TCG samples became darker, as is evident from the  $L^*$  coordinate value, where the lowest  $dL^*$  is G5 ( $dL^* = -0.99$ ) and the darkest is G30 ( $dL^* = -2.26$ ) compared to the controlled sample G0. Then, for  $a^*$  and  $b^*$  coordinates, all TCG samples exhibited positive values, signifying that the red and yellow tones appeared after plasma treatment, respectively. In addition, the values of  $a^*$  and  $b^*$  increased as the plasma treatment exposure time increased. Therefore, in this study, the increase in nitrogen plasma exposure time from 5 to 30 min can affect the colour of the treated surface, whereas a shorter exposure time leads to less changes in its  $a^*$  and  $b^*$  values. Compared to all TCG samples, G30 showed the highest values for  $a^*$  and  $b^*$ , which were 3.16 (red tone) and 11.96 (yellow tone), respectively. Regarding the WI, UCG (G0) had the highest WI with 87.51, while G30, as expected, had the lowest WI with 79.93. This is because the appearance of  $a^*$  (red) and  $b^*$  (yellow) coordinates and surface oxidation resulting from plasma treatment significantly reduced the WI of G30.

The changes of  $a^*$  and  $b^*$  values in all TCG samples can be explained by thermal oxidation that occurs on cotton surface, which induces a certain degree of yellowness and redness on the treated surface and reduces the whiteness intensity (Kan and Lam 2018). Upon plasma treatment, the reactive plasma molecules carry high kinetic energy, eventually generating heat that can be transmitted to the cotton surface, causing the yellowing effect on the cellulosic chain of cotton fabric (Yang *et al.* 2010). Hence, a prolonged plasma exposure period will transmit more heat to the cotton surface, hence increasing the intensity of the yellowish tone and reducing its WI. In addition, the yellowness value in plasma-treated cotton is also due to the etched areas, which result in

high surface roughness. The reflection of light from smooth surfaces is greater than rough ones (Eyupoglu *et al.* 2015); therefore, the higher surface roughness due to prolonged plasma treatment will reflect less light, proportionally increasing the yellowness value and decreasing the WI.

## CONCLUSIONS

As the plasma exposure time increased from 5 to 20 min, the TCG surface experienced increased surface roughness due to the etching effect and also experienced changes in surface chemical composition and surface charge, which turned the TCG into a cationic substrate with a large surface area. Plasma treatment from 5 to 20 min increased the percentage of CI and reduced the WI due to the etching effect and surface oxidation, respectively. Then, at 30 min, nitrogen plasma treatment on TCG started to show an early sign of degradation by experiencing a decrease in CI percentage, surface chemical functional group intensity, and surface charge. Plasma treatment at 20 min is considered an ideal exposure time where it has shown the optimum value for all the measured parameters, thus altering the cotton surfaces' functionally without affecting its bulk properties.

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