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ORIGINAL ARTICLE

Cotton textile with citronella nanoparticles: antimicrobial properties and surface functionalization strategies

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Abstract

The development of functional textiles offers wide-ranging applications, with nanoencapsulation of essential oils emerging as a promising strategy for antimicrobial purposes. This study focused on biodegradable nanoparticles loaded with citronella essential oil (CEO), deposited on cotton textiles using three methods, with or without non-thermal plasma (NTP) treatment. The immersion method achieved the highest CEO content per textile area (5.9 μL cm $^{-2}$). XPS and FT-IR analyses revealed that NTP treatment enhanced the hydrophilic functional groups through oxidation, as confirmed by contact angle assays and textile mass loss. Despite these changes, NTP treatment did not significantly alter the in vitro release profile of the CEO. Non-treated NTP samples were tested against *S. aureus* and *P. aeruginosa*, showing a stronger antibacterial effect against gram-positive *S. aureus.* These findings highlight the potential of these materials for use as functional antibacterial textiles with promising applications in health and hygiene products.

Keywords: *biofunctional textiles, essential oil release, non-thermal plasma.*

Data Availability: All data supporting the findings of this study are included in this article and its supplementary materials.

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1. Introduction

New technologies and high demand for innovative products and applications affect the textile industry. In parallel, textiles, especially those made from natural fibers, are suitable substrates for propagating microorganisms such as fungi and bacteria, resulting in functional and visual alterations, including fiber damage, unpleasant odors, and hygiene issues[1-3]. In both contexts, biofunctional textiles with antimicrobial finishing are receiving significant attention as an advanced material for preventing bacterial infections[4]. Functionalized substrates include the term "biofunctional", which refers to materials that exert biological activity in the context of textiles. This advanced material contains active substances incorporated in drug delivery systems that allow a slow release of the compounds when rubbed on the skin[5-7]. A biofunctional textile can provide antimicrobial, antiperspirant, insect-repellent, and cosmetic functions. The non-clothing sector has also benefited from biofunctional textiles used to treat skin wounds[8].

Natural compounds such as essential oils have been widely reported regarding antimicrobial activities[9,10]. However, due to their high vapor pressure, the encapsulation of essential oils has been suggested to preserve their biological features. Polymeric nanoparticles are often used for encapsulation purposes, where chitosan is commonly seen as a wall material in encapsulation methodology for essential oils[11]. Bioactivity, nontoxicity, biocompatibility, and antimicrobial properties are favored by reactive amino and hydroxyl groups along the polysaccharide backbone[12]. This polymer possesses attractive properties, making it a candidate to substitute synthetic polymers in textile fields[13].

Methods are also used to ensure the adhesion of micro- and nanoparticles to textile fibers. Fiedler et al.[14] used acid tetracarboxylic acid to functionalize non-woven cotton and promote cross-linking between starch microcapsules and the cotton. To obtain a functional textile through microencapsulation of citronella oil, Tariq et al.[15] employed gelatin and Gum Arabic as shell materials and an acrylic-based binder to fix the microparticles on the textile. As a promising alternative to the use of chemical agents for surface material functionalization, non-thermal plasma (NTP) treatment is widely discussed in the literature. The focus is on the production of radicals that increase hydrophilicity and micro-roughness, as well as a cleaning effect for textile surfaces, especially for cotton[16-19]. Naebe et al.[20] reported an improvement in chitosan adherence in cotton treated with O_2 plasma, justifying the occurrence of physical adsorption.

Through the plasma treatment, the oxygenated groups generated on the surface, such as C=O and C-O-C, act as binding sites on cotton, improving the adhesion between the textile and the polymer. Plasma treatment is an environmentally friendly option, using reagents that promote crosslinking between the substrate and the adsorbate.

In this context, the present research reports the bactericidal activities of biofunctionalized cotton textiles. The impregnated nanostructured system consisted of Pluronic F127 nanoparticles loaded with citronella essential oil and covered with a polyelectrolyte complex formed from chitosan and sodium alginate. The preparation and physicochemical characterization of nanoparticles were previously reported by our research group in Sanches et al.[11]. Three different impregnation methodologies were evaluated, and we investigated whether the plasma surface treatment of the textile was relevant for nanoparticles adhesion.

2. Materials and Methods

2.1 Materials

Nanoparticles impregnated in textiles were prepared and characterized, following the methodology proposed in a previous study reported by Sanches et al.[11]. This consists of citronella essential oil (Harmonia Natural) encapsulated in Pluronic® F127 (Sigma-Aldrich) micelles, covered by an electrolyte complex formed by medium molar mass chitosan (87% degree of deacetylation), viscosimetric molar mass (Mv) = 10.6×10^4 g mol $^{-1}$ and sodium alginate, Mv = 31.1×10^4 g mol $^{-1}$, both purchased from Sigma-Aldrich.

Cotton samples, used in the manufacture of the inner lining of children's shoes, were kindly donated by the company Contramão Calçados (São João Batista, SC, Brazil). To whiten the cotton, samples were washed in water/ NaClO (90:10 v/v). That is, to remove dirt that would obscure the characterization and nanoparticles impregnation. All the tests were conducted with washed samples.

2.2 Non-thermal plasma treatment for textiles

The textile treatment with NTP was made in a plasma reactor (Figure S1, Supplementary Material), using 1.0 mL/min oxygen flow, a 1 Torr pressure, and 7.4 W of power. The gap and dielectric barrier were fixed at 10.0 and 2.0 mm, respectively. The cotton textile sample (2.5 × 2.5 cm) was fixed in a borosilicate dish, centered on the bottom electrode, and exposed to a plasma glow discharge for 1 and 5 min (Cot-1 and Cot-5).

2.3 Contact angle and gravimetric analysis

The cotton wettability was observed using contact angle analysis (Θ) obtained with a Dataphysis goniometer equipped with a high-resolution camera, as it approached pure water. Four water drops (5.0 µL) were applied to the textile surface, in different positions, and processed automatically by SCA 20 software. In addition, the textiles were weighed before and after the process to evaluate the mass loss due to NTP surface treatment. The surface textile loss of weight was quantified using Equation 1, which calculates mass loss as:

$$Loss\ weight\,(\%) = \frac{M1 - M2}{M1} x100 \quad (1)$$

where M_1 and M_2 are the masses before and after treatment.

2.4 Fourier Transform Infrared Spectroscopy (FT-IR)

The white cotton samples that had been untreated and treated by non-thermal plasma were analyzed by FT-IR spectroscopy. The analyses were performed with a Shimadzu Prestige-21, equipped with an Attenuated Total reflection (ATR) accessory, and the data were collected over the range of 4000- 700 cm^{-1} with a resolution of 2.0 cm^{-1} and 25 scans.

2.5 X-ray photoelectron spectroscopy

The high-resolution spectra of O 1s and C 1s photoelectron lines were taken for the blank textile (Cot-n) and the textile treated with NTP Cot-1 and Cot-5, without any additional sample preparation. All analyses were performed as received i.e., without any additional sample preparation. In the case of all samples, O and C were the dominant features observed.

The composition analyses were performed based on the intensities of O 1s and C 1s in a standard way, using appropriate atomic sensitivity factors (ASFs). For analysis of the peak profiles, pseudo-Voigt GL(30) was used, and the constraint that FWHMs of all contributions in the frame of a line are the same for each C 1s and O 1s. The calibration was performed based on the position of C 1s, assuming this is situated at the characteristic position at 284.5 eV. Analyses were made in Multitechnical equipment, equipped with a VSW XPS system, which uses a non-monochromatic Mg Kα line, in FAT mode using 44 eV (survey) and 22 eV (high resolution)[21].

2.6 Nanoparticles impregnation in cotton textile

As previously mentioned, the aim was to achieve a bactericidal property in cotton textiles by impregnating citronella essential oil encapsulated into polymeric nanoparticles. This nanostructured system formulation was reported by Sanches et al.[11]. A representative measurement indicated a particle size of approximately 200 nm, PDI of 0.4, zeta potential of +50 mV, and the encapsulation efficiency was approximately 80%. To offer different methodologies of nanoparticles impregnation, three distinct processes are presented here. The first methodology consisted of textile immersion (IM1) in a nanoparticles dispersion in which textile samples (2.5 × 2.5 cm) were immersed in a 10 mL nanoparticles dispersion for 12 hours. The second (IM2) and third (IM3) methods were based on sprinkling NPs on the cotton textile under a vacuum system. Figure S2a represents the scheme of method IM2, where the sprinkling was done using a dripper (Razel (Georgia, Vermont) Modelo R-100EC) to control the flux (300 mL h $^{-1}$), a springer with a needle of 0.8 mm in diameter, and N_2 flux of 3.25 mL min $^{-1}$. Ten mL of NP suspension was previously filtered using a 1.2 µm filter and sprinkled onto a cotton fiber diameter of 1 cm.

In IM3 represented in Figure S2b, 10 mL of NPs suspension was sprinkled on a cotton textile sample with 1 cm of diameter, using an airbrush with an air flux of 4 mL min $^{-1}$. Samples after sprinkling processes by methods IM2 and IM3 were subsequently dried in a desiccator until the next analysis.

2.6 Scanning Electron Microscopy

The morphology of the textile samples was observed using a JEOL JSM-6390LV Scanning Electron Microscope at an accelerating voltage of 10 kV and 20 kV. A cotton piece of 0.5x0.5 cm was stuck on the stub and covered with gold.

2.7 The CEO loaded and in vitro release study

To find the CEO concentration per cm^2, three textile pieces of 2.5 × 2.5 cm were immersed in 120 mL of NP dispersion for 12 hours, with a CEO concentration of 14 uL mL^{-1}. After that, the cotton textile was gently dried with a paper towel to remove the excess dispersion and placed in 10 mL of hexane to extract the CEO content from the textile.

Assays of *in vitro* release of impregnated cotton textile (2.5 × 2.5 cm) were conducted in simulated sweat fluid (20 mL, pH = 4.3) at 37 °C in the water bath[22] with treated and non-treated NTP cotton textiles. All CEO quantifications were performed by UV-VIS spectrophotometry technique at 210 nm (UV NOVA/1800, Brazil). Periodically, aliquots of 3 mL were withdrawn to measure the absorbance. The CEO content (µL mL^{-1}) was estimated using a calibration curve of CEO in sweat fluid (y = 0.02867x + 18.174), with a coefficient of determination (R^2) of 0.9857, indicating an excellent linear correlation between the absorbance and CEO concentration.

2.8 Antimicrobial activity

Antimicrobial activity was evaluated for samples without plasma treatment, whose nanoparticles were impregnated by immersion. Tests were conducted according to the Technical Norm ABNT NBR 15275:2016 - Biological Assays, Insole, Synthetic Laminate, and Sole - Determination of resistance of microbial attack. Bactericidal action was evaluated against *Pseudomonas aeruginosa* (ATCC 24853) and *Staphylococcus aureus* (ATCC 6538), respectively, in concentrations of $4.6x10^7$ and $5.6x10^7$ cell mL^{-1}. After the incubation period for the strains, a quantity of each strain was spread on the TSB agar plate. A textile sample (2.5 × 2.5 cm) was rubbed to trigger citronella release and adhered to the agar plate. Tests were conducted in duplicate for each culture, and for the blank textile, that is, without nanoparticles.

3. Results and Discussions

3.1 Contact angle and gravimetric analysis

The contact angle of 141.5° ± 2.12 for untreated cotton indicates a highly hydrophobic surface, attributed to non-cellulosic components such as greases and pectins (0.4-1.2%)[23]. As shown in Figure S3, water droplets remain cohesive on the surface, suggesting weak adhesive interaction with the fabric. After oxygen plasma exposure, contact angle measurement was unfeasible due to immediate water absorption. The sample treated for 1 minute showed slower absorption, while the 5-minute treatment led to instantaneous absorption. This behavior is explained by the action of O_2 plasma filaments, which enhance cleaning and remove the hydrophobic cuticle, thereby increasing surface wettability and roughness[24,25].

According to Pandiyaraj and Selvarajan[26], plasma treatment alters the distribution of gaps between fibers and threads, promoting high liquid capillarity due to surface cleaning. The energy species generated during plasma discharge not only remove surface contaminants but also erode the cotton cuticle. This erosion was quantified using Equation 1, and, after 1 minute of treatment, a 3.10% mass loss was observed, increasing to 22.92% after 5 minutes, indicating that longer plasma exposure causes significant damage by removing not only the cuticle but also inner fiber layers. This trend reflects the sequential removal of surface additives, such as finishing agents. Therefore, while oxygen plasma is effective for surface cleaning, short treatment durations are recommended to preserve the structural integrity of the cotton substrate.

3.1 X-ray photoelectron spectroscopy

The results of general composition and fitting regarding oxygen 1s lines and carbon are shown in Table S1. Several conclusions can be extracted from the detailed analysis of the high-resolution spectra of C 1s and O 1s lines taken from cotton samples. The analysis related to the total composition of each element shows that for Cot-1, there was an increase of about 10% in groups containing oxygen, including possible contamination. For Cot-5, exposed to discharge for a longer time, the increase in oxygenated groups was not as pronounced.

The interpretation of O 1s contributions is not straightforward, but they surely correspond to different kinds of C-O bonds. All O 1s lines can be fitted to three contributions. Very high consistency between all samples concerning the positions of the three contributions can be observed. The O1 position refers to the C=O or O-(C=O*)-C groups in aliphatic hydrocarbons; the O2 position suggests correspondence with O-C-O or O*-(C=O)-C, and the O3 contribution is related to C-OH and C-O. Considering only the samples exposed to NTP, it seems evident that with 1 min of exposure, the O3 groups are initially formed, and at 5 min, these groups are partially oxidized to O1 and O2. It is also important to note that initially, these groups are present on the surface, either as a natural protective layer or as synthetic gum. Plasma acts by sputtering and oxidizing the innermost layers of cotton.

It can be seen that the standard C-C/C-H signal (C1) in the sample Cotton 1 showed low relative intensity, and as the sample is exposed to NTP, the signal for these species increased. This event may be related to the presence of fragments due to oxidative degradation promoted by NTP or the exposure of natural fibers to plasma. The C-OH/C-O species, represented by the C2 peak, showed a significant decrease in intensity when compared to the Cot-n sample, suggesting that these species may be being converted into groups with a higher oxidation state (C3 and C4), as can be observed in Table S1.

There was a clear increase in the relative intensity of the peaks related to the O-C-O (C3) groups with exposure to plasma, justifying the oxidation of the sample. There was, however, a fourth contribution, represented by C4, near 290 eV, which also increases according to NTP exposure, whose displacement (with charging correction) is characteristic of groups O-C=O.

3.2 Fourier Transform Infrared Spectroscopy

Non-treated samples and oxygen NTP-treated cotton textiles were compared regarding their functional groups, as can be seen in Figure S4. The external layer of cotton fiber consists mostly of pectin; the band at 1717 cm $^{-1}$ is assigned to the group carboxyl of this polymer, and the relative intensity in the ALG-ST sample is pronounced. For the Cot-1 and Cot-5 samples, it is noted that, as the time of exposure to the plasma increases, the band at 1717 cm $^{-1}$ remains intense. It can be associated with the hemicellulose groups from the primary and secondary walls of the cotton fiber that were exposed due to plasma treatment. The band is 1409 cm $^{-1}$, corresponding to the vibration of symmetric folding in the plane of the C-H connection[27]. According to Meyabadi[28], 1409 cm $^{-1}$ is a band of the crystalline region of the material, whose relative intensity in the present work is amplified in samples with longer surface treatment time. The bands at 1338 and 1250 cm^{-1} are referring to out-of-plane bending vibrations of the C-H linkage. The band at 1120 cm^{-1} refers to the asymmetric stretch vibration of the C-O-C bond of the glycosidic ether, due to the content of cellulosic material[29]. The band at 1018 cm^{-1} is associated with the stretch of the C-O linkage. All bands are evidenced with increasing exposure time from the sample to the plasma, again indicating that this surface treatment degrades first the cuticle, which is the outer layer composed of greases and pectins, and then the primary wall of the cellulose, exposing the other groups of hydrophilic character, which are under the XPS results.

3.3 Scanning Electron Microscopy

SEM images were obtained for bleached cotton before and after NTP exposure. In Figure 1a the textile surface is smooth with no defects.

The yarns are still covered with the cuticle, whose layers are responsible for the hydrophobic character of the fabric, in agreement with the high contact angle values found for this sample. After exposure to NTP, the surface morphology is visually changed, and as observed in the SEM images, it is dependent on the treatment time. In Figure 1b, the sample that was exposed to 5 minutes of NTP treatment resulted in an increase in roughness. The removal of the hydrophobic cuticle is observed due to the appearance of the longitudinal fibrils that characterize the primary wall of the cotton fiber, due to the short treatment time with plasma[30]. Besides the exposure of the fibrils, reactive oxygen species formed valleys on the fabric surface due to the bombardment of O_2-formed species.

In Figures S5a, S5b, S5c, and S5d, it is observed that NTP treatment even helps to increase the space between the threads and disarrange the fibers. These changes in morphology are also related to greater capillary flow observed in improved water absorption in contact angle measurements for samples that have been exposed to NTP as a function of different treatment times[30,31].

Similar results were obtained from SEM images in the work of Sarma et al.[32] and Prysiazhnyi et al.[33]. The authors found that the damage caused to the fiber structure is justified by the removal of the amorphous regions of the outer layer and, consequently, the attack of O_2 radical species in the crystalline region of the fiber. These observations regarding the morphology are in agreement with the suggestions in the analysis of the FT-IR spectra. They suggest an increase in the intensity of the functional groups with the time of treatment, due to the removal of the cuticle and exposure of the inner walls of the fiber.

3.4 Nanoparticles impregnation in cotton textile

Deposition and nanoparticles distribution in textile fibers were also observed, according to the method of impregnation used. Figure 2 refers to the cotton without NTP treatment, where in 2a the deposition was carried out by immersion in NPS suspension and in 2b by the IM2 method. In 2a, in addition to a few spherical agglomerates, there is a small film circled in black.

In 2b, it is possible to note the presence of spheres and other agglomerates that may be characterized as polymeric agglomerates originating from the nanoparticles. The adhesion of nanoparticles to the non-treated NTP fibers can be explained by the heterogeneous coating of their hydrophobic cuticle. The exposed pores of the fiber act as anchors for the adhesion of nanoparticles[33].

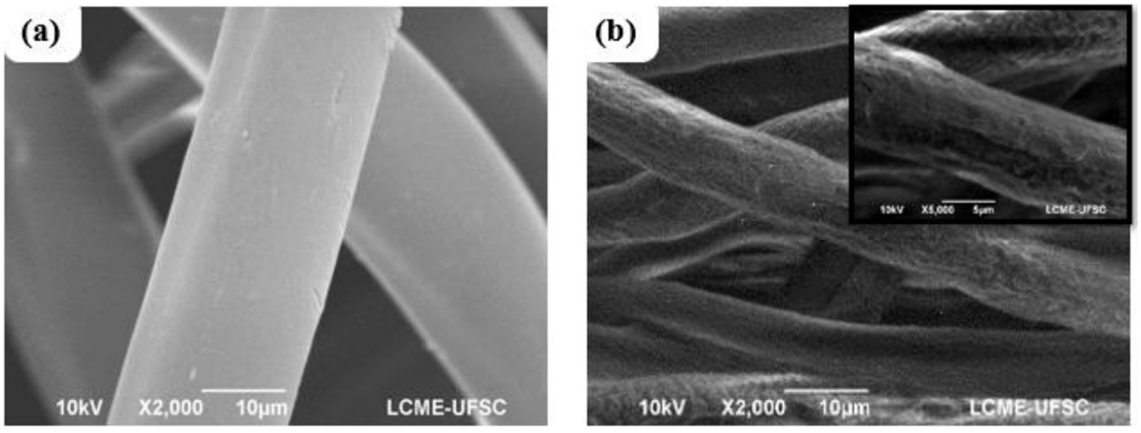

Figure 1. Cot-n (a) and Cot-5 (b).

Figure 3 exhibits the images of samples previously exposed to NTP, Cot-1, which received the deposition of the nanoparticles. In 3a and b, the exposure was by the immersion method, in 3c and 3d by the IM2, and in 3e and 3f by the IM3 method. Figure 3b shows small spheres adhered to the fiber. In images 3c and 3d, there are a large number of irregular agglomerates well dispersed in the fibers and with size polydispersity. Crater-like structures were observed on the surface of fibers (Figure 3d), which is commonly reported in the literature for cotton fabric after treatment with oxygen plasma[33].

In Figures 3e and 3f, it can be noted that the sprinkling methodology with no filtered samples and sprinkled at a high flow rate contributes to forming a film above the fibers. This is a non-required result from the point of view of the consumer experience since film formation could occlude the empty spaces in the textile, reducing the transpiration.

After 5 min of NPT and applying the IM1 method, the SEM images in Figures 3g and 3h were obtained. The results show irregular agglomerated structures adhered to the fiber's surface, making it possible to identify nanoparticles as observed for the samples in Figure 3.

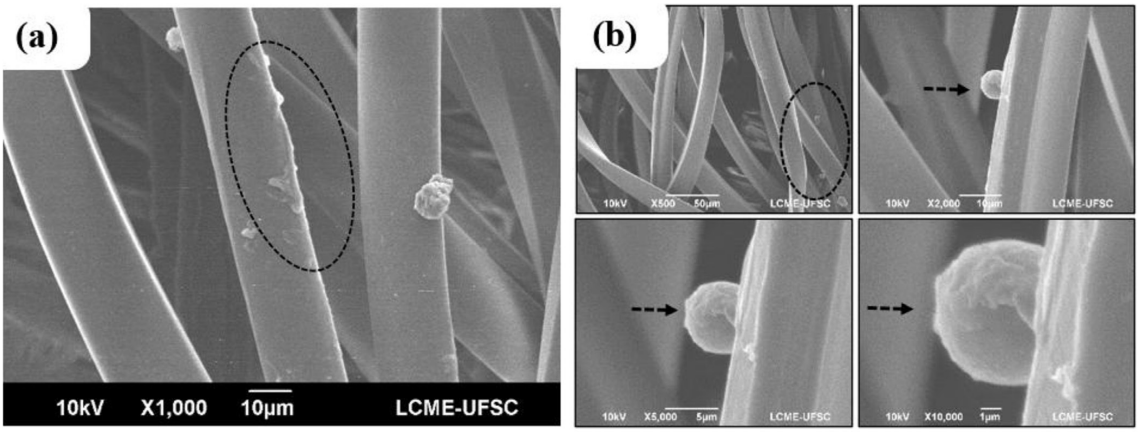

Figure 2. Cotton fibers not exposed to NTP after IM1 (a) and IM2 (b).

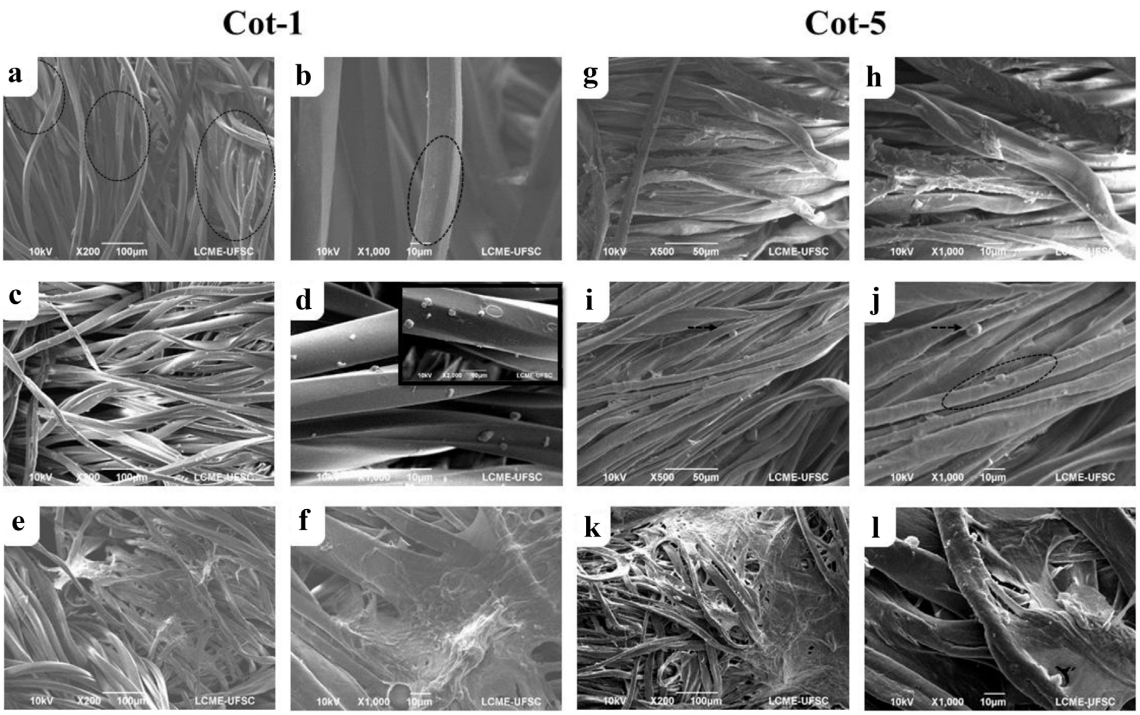

Figure 3. Cot-1 after impregnation methods IM1 (a) and (b), IM2 (c) and (d), and IM3 (e) and (f), and Cot-5, IM1 (g) and (h), IM2 (i) and (j), and IM3 (k) and (l).

For deposition via IM2 (Figure 3i and 3j), there is no observation of polymeric agglomerates covering the fibers, but in J, the fibers are twisted and open in the longitudinal direction, and there was deposition in this hollow, as indicated by black highlighting. As in sample Cot-1, 3e, and 3f images show the deposition of the same polymer material on the textile surface.

According to Fiedler et al.[14], microcapsules of smaller size facilitate adsorption and penetration into the fabric's surface due to the occupation of the interstices between the threads and fibers. Smaller-sized particles are also advantageous in controlling the dosage of actives and in fabric durability[34].

3.5 Essential oil in vitro release from cotton textile

The CEO content in the textile sample was measured by UV-Vis spectrophotometry, as described in the methodology section. For the IM1 proceeding, 5.9 μL cm $^{-2}$ was found, while for IM2 and IM3, the values were 3.95 and 4.47 μL cm $^{-2}$, respectively. The greater amount in IM1 is justified by the adsorption of free CEO, around 20% of the total amount, according to the results of EE%. The decrease in the total amount of CEO retained in the textile could be explained by the effect of the vacuum system applied in IM2 and IM3, which can contribute to vaporizing the non-loaded CEO.

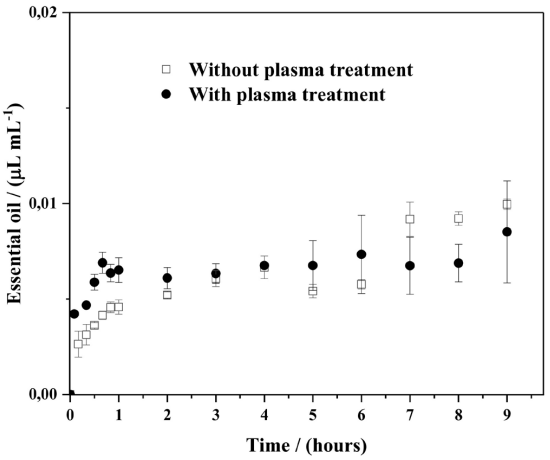

Figure 4. Release profile for untreated (▫) and treated (●) cotton fiber with non-thermal plasma, impregnated by the immersion method.

The *in vitro* release assays were performed only for the samples obtained by IM1, since those samples show the highest CEO content. Figure 4 exhibits two release curves from cotton samples impregnated with NPs via immersion for treated and non-treated textiles. Variance analysis shows that $F_{crit} > F_{calc}$ ($p=0.05$), so it can be suggested that the plasma surface treatment does not significantly improve the nanoparticles-textile interaction and, consequently, the release profile of CEO, under the studied conditions. For this reason, the non-treated cotton sample was chosen to perform the microbiological test.

3.6 Microbiologic activity from cotton textile

The microbiologic test, performed under static conditions, is based on the growth of the microorganism when it comes into close contact with the impregnated sample. On the other hand, the active compound is subjected to mechanisms of transport and diffusion from the textile to the plate, where its activity inhibits growth under and around the textile[35]. The bactericidal activity of the textile against the gram-negative bacteria *P. aeruginosa* is observed in Figures 5a, 5b, 5c, and 5d, and Figures 5e, 5f, 5g, and 5h, the result of the test performed for the gram-positive bacteria *S. aureus*. For blank samples, that is, without nanoparticles treatment, the growth of the colonies is observed in the form of fine films, visualized without the use of a microscope on the entire plate, including the underside of the fabric.

The bactericidal activity of cotton fabric treated with NPs loaded with CEO is shown in Figures 5c, 5d, 5g, and 5h, in which the final concentration of the active was about 5.9 ± 0.37 μL cm^2.

According to the standard ABNT NBR 15275, the evaluation is only related to the growth or no growth under de sample material. For example, the growth under the samples

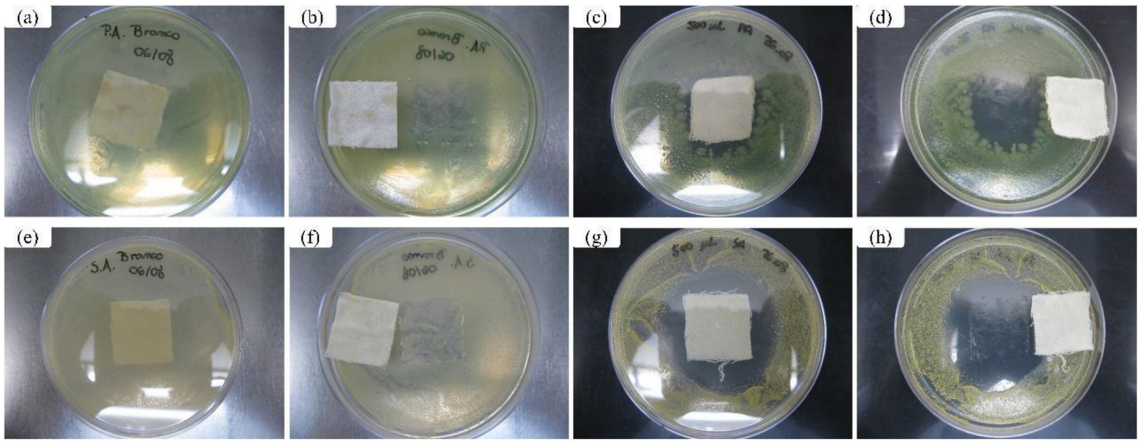

Figure 5. Blank textile (a) and (b); inhibition halo (c) and (d) for *P. aeruginosa.* Blank textile (e) and (f) and inhibition halo (g) and (h) for *S. aureus*.

was observed in Figures 5b and 5f, and in Figures 5d and 5h, there was a growth inhibition. Suppose there is an inhibition zone (halo) around the sample material. In that case, it means that the biocide reagent is soluble in water and that diffusion creates a concentration gradient of the biocide reagent around the sample material. If the halo is greater in the case of a specific bacteria, it means that lower active concentrations are sufficient to inhibit the growth of that organism, compared to another. In this context, the results have proven to be efficient in terms of bacterial resistance for the two organisms in the study.

Similar results were reported by Hidayah et al.[36], that there is a greater halo of inhibition against *S. aureus* when compared to *P. aeruginosa,* once citronella EO has bactericidal characteristics that inhibit the growth of *S. aureus* biofilm. Lopez-Romero et al.[37] attribute to citronellol and citronellal the bactericidal activity of both gram-positive and gram-negative bacteria. These terpenoids are capable of disrupting the hydrophilic channels in the outer membrane, even of gram-negative bacteria. Rodríguez-Lopez et al.[38] confirm the effect of monoterpenes from essential oils on the permeability of the outer membrane of gram-negative bacteria.

4. Conclusions

Treating the cotton surface with non-thermal oxygen plasma for 1 or 5 minutes did not significantly improve the adhesion of nanoparticles containing citronella essential oil (CEO). Despite the increase in wettability and chemical and morphological modification of the fibers after treatment, especially with 5 minutes of exposure, the data obtained did not indicate direct benefits in the loading or release of the active ingredient. Among the impregnation methods evaluated, immersion deposition stood out as the most efficient strategy for incorporating CEO. The sample functionalized by this method, without plasma pre-treatment, showed bactericidal activity against *S. aureus* and *P. aeruginosa*, with greater efficacy against gram-positive bacteria. The results reinforce the potential of using nanoparticles containing CEO in the antimicrobial functionalization of fabrics and highlight the viability of simple methods such as immersion for application to cellulose substrates.

5. Author's Contribution

- **Conceptualization** – Mariele Paludetto Sanches; Alexandre Luís Parize; Valdir Soldi
- **Data curation** – Mariele Paludetto Sanches; Rodrigo Henrique Saatkamp; Idejan Padilha Gross; Taís Felix
- **Formal analysis** – Mariele Paludetto Sanches; Rodrigo Henrique Saatkamp; Idejan Padilha Gross; Taís Felix; Markus Wilimzig
- **Funding acquisition –** Valdir Soldi; Alexandre Luís Parize.
- **Investigation** – Mariele Paludetto Sanches; Rodrigo Henrique Saatkamp; Idejan Padilha Gross; Taís Felix; Markus Wilimzig
- **Methodology** – Mariele Paludetto Sanches; Rodrigo Henrique Saatkamp; Idejan Padilha Gross; Taís Felix; Markus Wilimzig
- **Project administration** – Alexandre Luís Parize; Valdir Soldi
- **Resources** – Valdir Soldi; Alexandre Luís Parize; Nito Angelo Debacher
- **Software** – NA.
- **Supervision** – Alexandre Luís Parize; Nito Angelo Debacher; Valdir Soldi
- **Validation** – NA.
- **Visualization** – Mariele Paludetto Sanches; Rodrigo Henrique Saatkamp
- **Writing – original draft** – Mariele Paludetto Sanches; Rodrigo Henrique Saatkamp
- **Writing – review & editing** – Mariele Paludetto Sanches; Rodrigo Henrique Saatkamp; Alexandre Luís Parize

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7. References

1. Tanasa, F., Teaca, C.-A., Nechifor, M., Ignat, M., Duceac, I. A., & Ignat, L. (2023). Highly specialized textiles with antimicrobial functionality-advances and challenges. *Textiles*, *3*(2), 219-245. https://doi.org/10.3390/textiles3020015.
2. Gulati, R., Sharma, S., & Sharma, R. K. (2022). Antimicrobial textile: recent developments and functional perspective. *Polymer Bulletin*, *79*(8), 5747-5771. https://doi.org/10.1007/s00289-021-03826-3. PMid:34276116.
3. Broadhead, R., Craeye, L., & Callewaert, C. (2021). The future of functional clothing for an improved skin and textile microbiome relationship. *Microorganisms*, *9*(6), 1192. https://doi.org/10.3390/microorganisms9061192. PMid:34073029.
4. Oliveira, C. S. F., & Tavaria, F. K. (2023). The impact of bioactive textiles on human skin microbiota. *European Journal of Pharmaceutics and Biopharmaceutics*, *188*, 66-77. https://doi.org/10.1016/j.ejpb.2023.05.004. PMid:37182552.
5. Massella, D., Argenziano, M., Ferri, A., Guan, J., Giraud, S., Cavalli, R., Barresi, A. A., & Salaun, F. (2019). Bio-functional textiles: combining pharmaceutical nanocarriers with fibrous materials for innovative dermatological therapies. *Pharmaceutics*, *11*(8), 403. https://doi.org/10.3390/pharmaceutics11080403. PMid:31405229.
6. Repon, M. R., Islam, T., Islam, T., Ghorab, A. E., & Rahman, M. M. (2023). Cleaner pathway for developing bioactive textile materials using natural dyes: a review. *Environmental Science and Pollution Research International*, *30*(17), 48793-48823. https://doi.org/10.1007/s11356-023-26131-0. PMid:36879092.

7. Freitas, A. S., Oliveira, R., Ribeiro, A., & Almeida-Aguiar, C. (2024). Biofunctional textiles: antioxidant and antibacterial finishings of cotton with propolis and honey. *International Journal of Molecular Sciences*, *25*(15), 8034. https://doi.org/10.3390/ijms25158034. PMid:39125604.
8. Shah, M. A., Pirzada, B. M., Price, G., Shibiru, A. L., & Qurashi, A. (2022). Applications of nanotechnology in smart textile industry: a critical review. *Journal of Advanced Research*, *38*, 55-75. https://doi.org/10.1016/j.jare.2022.01.008. PMid:35572402.
9. Kant, R., & Kumar, A. (2022). Review on essential oil extraction from aromatic and medicinal plants: techniques, performance and economic analysis. *Sustainable Chemistry and Pharmacy*, *30*, 100829. https://doi.org/10.1016/j.scp.2022.100829.
10. Coimbra, A., Ferreira, S., & Duarte, A. P. (2022). Biological properties of *Thymus zygis* essential oil with emphasis on antimicrobial activity and food application. *Food Chemistry*, *393*, 133370. https://doi.org/10.1016/j.foodchem.2022.133370. PMid:35667177.
11. Sanches, M. P., Gross, I. P., Saatkamp, R. H., Parize, A. L., & Soldi, V. (2020). Chitosan-sodium alginate polyelectrolyte complex coating Pluronic® F127 nanoparticles. *Journal of the Brazilian Chemical Society*, *31*(4), 803-812. https://doi.org/10.21577/0103-5053.20190244.
12. Yan, D., Li, Y., Liu, Y., Li, N., Zhang, X., & Yan, C. (2021). Antimicrobial properties of chitosan and chitosan derivatives in the treatment of enteric infections. *Molecules*, *26*(23), 7136. https://doi.org/10.3390/molecules26237136. PMid:34885715.
13. Li, J., Fu, J., Tian, X., Hua, T., Poon, T., Koo, M., & Chan, W. (2022). Characteristics of chitosan fiber and their effects towards improvement of antibacterial activity. *Carbohydrate Polymers*, *280*, 119031. https://doi.org/10.1016/j.carbpol.2021.119031. PMid:35027133.
14. Fiedler, J. O., Carmona, O. G., Carmona, C. G., Lis, M. J., Plath, A. M. S., Samulewski, R. B., & Bezerra, F. M. (2019). Application of *Aloe vera* microcapsules in cotton nonwovens to obtain biofunctional textiles. *Journal of the Textile Institute*, *111*(1), 68-74. https://doi.org/10.1080/00405000.2019.1625607.
15. Tariq, Z., Izhar, F., Gmd, Z., Zulfiqar, A., Malik, M. H., Oneeb, M., & Khan, A. (2022). Fabrication of higly durable functional textiles through microencapsulation of organic citronella oil. *Industrial Crops and Products*, *190*, 115878. https://doi.org/10.1016/j.indcrop.2022.115878.
16. Cruz, J., & Fangueiro, R. (2016). Surface modification of natural fibers: a review. *Procedia Engineering*, *155*, 285-288. https://doi.org/10.1016/j.proeng.2016.08.030.
17. Kert, M., Tavcer, P. F., Hladnik, A., Spasić, K., Puač, N., Petrović, Z. L., & Gorjanc, M. (2021). Application of fragrance microcapsules onto cotton fabric after treatment with oxygen and nitrogen plasma. *Coatings*, *11*(10), 1181. https://doi.org/10.3390/coatings11101181.
18. Zahid, M., Mazzon, G., Athanassiou, A., & Bayer, I. S. (2019). Environmentally benign non-wettable textile treatments: a review of recent state-of-the-art. *Advances in Colloid and Interface Science*, *270*, 216-250. https://doi.org/10.1016/j.cis.2019.06.001. PMid:31277037.
19. Ssekasamba, H., Tebyetekerwa, M., Haodong, C., Balilonda, A., Ssegwanyi, S., Gao, Q., & Xiaoliang, T. (2024). Review of plasma surface engineering technology toward sustainable textile materials. *Materials Circular Economy*, *6*(1), 27. https://doi.org/10.1007/s42824-024-00114-z.
20. Naebe, M., Li, Q., Onur, A., & Denning, R. (2016). Investigation of chitosan adsorption onto cotton fabric with atmospheric helium/oxygen plasma pre-treatment. *Cellulose*, *23*(3), 2129-2142. https://doi.org/10.1007/s10570-016-0915-0.
21. Felix, T., Cassini, F. A., Benetoli, L. O. B., Dotto, M. E. R., & Debacher, N. A. (2017). Morphological study of Polymer surfaces exposed to non-thermal plasma based on contact angle and the use of scaling laws. *Applied Surface Science*, *403*, 57-61. https://doi.org/10.1016/j.apsusc.2017.01.036.
22. Kulthong, K., Srisung, S., Boonpavanitchakul, K., Kangwansupamonkon, W., & Maniratanachote, R. (2010). Determination of silver nanoparticle release from antibacterial fabrics into artificial sweat. *Particle and Fibre Toxicology*, *7*(1), 8. https://doi.org/10.1186/1743-8977-7-8. PMid:20359338.
23. Ali, Z., Talpur, F. N., Afridi, H. I., Ahmed, F., Brohi, N. A., & Abbasi, H. (2025). Analytical approaches and advancement in the analysis of natural and synthetic fiber: a comprehensive review. *Spectrochimica Acta. Part A, Molecular and Biomolecular Spectroscopy*, *326*, 125164. https://doi.org/10.1016/j.saa.2024.125164. PMid:39332174.
24. Takahashi, G., & Akashi, H. (2011). Various kinds of streamers in atmospheric pressure oxygen dielectric barrier discharge. *IEEE Transactions on Plasma Science*, *39*(11), 2234-2235. https://doi.org/10.1109/TPS.2011.2160564.
25. Kan, C.-W., & Man, W.-S. (2018). Surface characterization of atmospheric pressure plasma-treated cotton fabric -effect of operation parameters. *Polymers*, *10*(3), 250. https://doi.org/10.3390/polym10030250. PMid:30966285.
26. Pandiyaraj, K. N., & Selvarajan, V. (2008). Non-thermal plasma treatment for hydrophilicity improvement of grey cotton fabrics. *Journal of Materials Processing Technology*, *199*(1-3), 130-139. https://doi.org/10.1016/j.jmatprotec.2007.07.046.
27. Vestena, M., Gross, I. P., Muller, C. M. O., & Pires, A. T. N. (2016). Isolation of whiskers from natural sources and their dispersed in a non-aqueous medium. *Polímeros: Ciência e Tecnologia*, *26*(4), 327-335. https://doi.org/10.1590/0104-1428.2367.
28. Meyabadi, T. F., Dadashian, F., Sadeghi, G. M. M., & Asl, H. E. Z. (2014). Spherical cellulose nanoparticles prepatation from waste cotton using a green method. *Powder Technology*, *261*, 232-240. https://doi.org/10.1016/j.powtec.2014.04.039.
29. Li, M., He, B., Chen, Y., & Zhao, L. (2021). Physicochemical properties of nanocellulose isolated from cotton stalk waste. *ACS Omega*, *6*(39), 25162-25169. https://doi.org/10.1021/acsomega.1c02568. PMid:34632175.
30. Hossain, M. M., Herrmann, A. S., & Hegemann, D. (2006). Plasma hydrophilization effect on different textile structures. *Plasma Processes and Polymers*, *3*(3), 299-307. https://doi.org/10.1002/ppap.200500152.
31. Man, W. S., Kan, C. W., & Ng, S. P. (2014). The use of atmospheric pressure plasma treatment on enhancing the pigment application to cotton fabric. *Vacuum*, *99*, 7-11. https://doi.org/10.1016/j.vacuum.2013.04.018.
32. Sarma, B., Sarma, A., & Rani, K. V. (2018). Plasma treatment on cotton fabrics to enhance the adhesion of reduced graphene oxide for electro-conductive properties. *Diamond and Related Materials*, *84*, 77-85. https://doi.org/10.1016/j.diamond.2018.03.009.
33. Prysiazhnyi, V., Kramar, A., Dojcinovic, B., Zekic, A., Obradovic, B. M., Kuraica, M. M., & Kostic, M. (2013). Silver incorporation on viscose and cotton fibers after air, nitrogen and oxygen DBD plasma pretreatment. *Cellulose*, *20*(1), 315-325. https://doi.org/10.1007/s10570-012-9817-y.
34. Li, W.-R., Shi, Q.-S., Ouyang, Y.-S., Chen, Y.-B., & Duan, S.-S. (2013). Antifungal effects of citronella oil against *Aspergillus niger* ATCC 16404. *Applied Microbiology and Biotechnology*, *97*(16), 7483-7492. https://doi.org/10.1007/s00253-012-4460-y. PMid:23081773.
35. López, A., Lis, M. J., Bezerra, F. M., Vilaseca, M., Vallés, B., Prieto, R., & Simó, M. (2019). Production and evaluation of antimicrobial microcapsules with essential oils using complex coacervation. *Journal of Biomedical Science and Engineering*, *12*(8), 377-390. https://doi.org/10.4236/jbise.2019.128029.

36. Hidayah, A. N., Wasito, E. B., Debora, K., Basori, A., Isnaeni, I., & Utomo, B. (2019). Correlation between the bacteriostatic and bactericide effect with antibiofilm and anticolony spreading from Javanese Citronella oil on methicillin-resistant *Staphylococcus aureus* (MRSA). *Folia Medica Indonesiana*, *55*(1), 1-9. https://doi.org/10.20473/fmi.v55i1.12542.
37. Lopez-Romero, J. C., González-Ríos, H., Borges, A., & Simões, M. (2015). Antibacterial effects and mode of action os selected essential oils components against *Escherichia coli* and *Staphylococcus aureus. Evidence-Based Complementary and Alternative Medicine: eCAM*, *2015*(1), 795435. https://doi.org/10.1155/2015/795435. PMid:26221178.
38. Rodríguez-Lopez, M. I., Mercader-Ros, M. T., Pellicer, J. A., Gómez-López, V. M., Martínez-Romero, D., Núñez-Delicado, E., & Gabaldón, J. A. (2020). Evaluation of monoterpene-cyclodextrin complexes as bacterial growth effective hurdles. *Food Control*, *108*, 106814. https://doi.org/10.1016/j.foodcont.2019.106814.

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Supplementary Material

Supplementary material accompanies this paper.

Table S1. Summary of cotton sample compositions by X-ray photoelectron spectroscopy.

Figure S1. Non-thermal plasma reactor scheme: a) power supply, b) electrodes, c) dielectric barrier, d) sample, e) gas inlet, and f) vacuum pump.

Figure S2. Scheme of impregnation methods: (a) spray I and (b) spray II. Legend: a) compressor air, a') N2 compressed gas, b) airbrush, b') Springer with a needle, c) NPs spray, d) cotton textile support, e) vacuum pump, f) zoom of cotton textile fixed on support, and g) dripper.

Figure S3. Contact angle analysis of an untreated textile with water.

Figure S4. Infrared spectra curves of cotton fiber untreated and NTP treatment.

Figure S5. Cot-n (a), Cot-1 (b) and (c), and Cot-5 (d).

Figure S6. Calibration curve of citronella essential oil in simulated sweat fluid, pH=4.3.

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