

COLD PLASMA TREATMENT OF COTTON AND VISCOSE FABRICS
IMPREGNATED WITH ESSENTIAL OILS OF
LAVANDULA ANGUSTIFOLIA AND *MELALEUCA ALTERNIFOLIA*

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*Dedicated to the 50th anniversary
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In this research, the synergetic effect of cold plasma treatment and two essential oils, namely *Lavandula angustifolia* (lavender) and *Melaleuca alternifolia* (tea tree), on the physicochemical properties of cotton and viscose fabrics were investigated. The chemical changes on the fabric surface caused by the treatments were investigated by contact angle measurements, surface energy calculation and ATR-FTIR analysis. In addition, the content of carboxyl, aldehyde and hydroxyl end groups was assessed. The antimicrobial activity of the untreated and treated fabrics was evaluated. The results showed that grafting with monomers existing in lavender or tea tree oil leads to an increase of the water contact angle and its stability over time. It was observed that the treatments with essential oils increase the carboxyl groups content, except for the viscose treated with lavender oil. It was also demonstrated that the cotton fabric immersed into lavender oil exhibited antimicrobial activity against *S. aureus*. However, further plasma treatment resulted in the loss of antibacterial properties.

Keywords: *Lavandula angustifolia*, *Melaleuca alternifolia*, cold plasma, textile fabrics, physicochemical properties, antimicrobial activity.

INTRODUCTION

Nosocomial infection currently represents not only a serious concern to the competent health agencies, but also a problem with social, ethical and legal implications in relation to the lives of patients. In order to reduce infection rates, many strategies and different methods have been developed in the last several decades.¹

Improved hygiene and healthcare standards have a great impact on the development of hygiene and health care products. Therefore, textiles related to health care have started receiving much attention, thus increasing the need for antimicrobial treatment of fabrics.^{2,3}

Cotton and viscose fabrics, in addition to being used in everyday clothing, are also applied in the medical and technical textile field.³⁻⁷ However, these cellulosic textiles are susceptible to the

microbial attack during use and storage. The growth of microbes on textiles has the potential to cause cross infection, transfer of diseases, allergic reactions, and unpleasant odours on humans.⁸ These detrimental effects can be controlled by antimicrobial finishing of the textile fabrics, which includes treatment with nano-scale metal or metal oxides, quaternary ammonium salt, antibiotics, natural extracts, essential oils or components of essential oils.^{2,5,6,9-13} The use of the latter has become popular, as numerous studies have been published on their antimicrobial activities against many different types of microorganisms.^{4,14-25}

Essential oils represent complex mixtures of chemical compounds (such as phenols, aldehydes, ketones, alcohols, esters, ethers or hydrocarbons)

with different antimicrobial properties, and for these reasons, it is very difficult to reduce their antimicrobial effect to one or several active principles.^{14,22} Studies developed by Jianu *et al.*¹⁹ and Soković *et al.*²² suggest that the components present in great proportions are not necessarily responsible for the total activity of the oil. The antibacterial activity exhibited by the oils can be explained by either the synergistic effect of the different components in the oil and/or by the presence of other components that may be active even in small concentrations.^{14,19,22} So, it is more meaningful and rational to study the whole essential oil rather than some of its components, also considering the synergism among the components in essential oils.¹⁴ The essential oils of *Lavandula angustifolia* and *Melaleuca alternifolia* are alcohol-based, exhibiting varying degrees of activity, depending on whether the strains are Gram-positive or Gram-negative.^{16,21,22} According to Mayaud *et al.*,²¹ of three alcohol-based essential oils tested (tea tree, palmarosa and lavender), tea tree oil was the most efficient against all tested Gram-negative bacteria, except *P. aeruginosa*. For lavender oil, the activity was better against Gram-positive than against Gram-negative bacteria. The compounds contained in lavender essential oil can vary over a considerable range, linalool (27.2-52.6%) and linalyl acetate (9.27-36.7%) being the major components.^{16,22,25} However, Van Vuuren *et al.*²⁵ also found a significant amount of terpinen-4-ol (14.9%), while Danh *et al.*¹⁶ found about 7% of both camphor and borneol. The variations in the composition of essential oils might arise from distinct extraction methods, environmental differences, such as climatic, seasonal or geographical conditions, and genetic differences. The different chemical compositions may have an impact on the antimicrobial activity.^{16,23} The antibacterial and antifungal properties of tea tree oils have been linked to their major component terpinen-4-ol.^{15,18} This component may represent about 34.8-49.3% of the oil, however other components, such as α - and γ -terpinene and cineole, are also present in considerable amounts.^{18,20,21,25}

Plasma treatment is a useful and suitable technique to modify a polymer surface, mainly of natural polymers, such as cellulose,^{2,4,5,7,9,10,12,26-30} or to polymerize organic molecules, which do not normally polymerize using conventional techniques.³¹⁻³⁷ Surface modification with plasma

can be used to incorporate functional properties, such as wettability, water repellency and antimicrobial finish.^{2,4-7,9-12,26-28,30} The production of antimicrobial organic plasma polymerized coatings has received a great deal of attention, in order to develop surfaces that prevent adhesion and proliferation of pathogenic bacteria.^{30-32,36} To the best of our knowledge, the plasma is applied as a pre-treatment of textile fabrics followed by antimicrobial treatment. In our work, the plasma treatment is made after an initial impregnation of the fabrics with essential oils, in order to modify the textile fabrics, to change their wettability and afford antimicrobial properties through polymerization of the compounds present in essential oils, assisted by cold plasma. Thus, cotton and viscose fabrics were subjected to three distinct treatments: cold plasma treatment, immersion into lavender or tea tree oil, and grafting with monomers existing in lavender or tea tree oil using cold plasma. The physicochemical properties, chemical nature and antimicrobial properties of the samples were evaluated.

EXPERIMENTAL

Materials

The fabrics used throughout this work were 100% viscose (CV) with a basis weight of 84 g/m² and thickness of 1.65 mm, and 100% cotton (CO) with a basis weight of 133 g/m² and thickness of 2.35 mm. Before the treatment, the samples were washed with 1 g/L nonionic detergent and dried in a laboratory oven. The fabrics were cut to dimensions of 3 cm x 5 cm. Two commercial essential oils were used: *Lavandula angustifolia* flower oil (LA) and *Melaleuca alternifolia* leaf oil (TT).

Plasma treatment

The radio frequency plasma generator used was a EUROPLASMA apparatus equipped with a microcontroller, a vacuum system and a 2.54 GHz microwave generator. The vacuum level was measured by a Pirani-type pressure gauge and the calibration was made using nitrogen. The reactor electrodes consisted of a cylindrical pyrex glass tube with a diameter and length of 60 mm. The energy input frequency was 13.56 MHz. The chamber was an aluminum made cylinder with a wall thickness of 2 cm. The useful dimensions of this cylindrical chamber were 200 and 150 mm, for the diameter and length, respectively. Some preliminary trials were carried out based on the experimental conditions described by Gaiolas *et al.*²⁸ Thus, the fabrics were impregnated with essential oils, followed by plasma activation and then again immersed into the oils for 20 minutes. The plasma

operating conditions were as follows: treatment power 200 W, constant pressure of 700 mTorr, and treatment times of 30 and 90 seconds, using argon as work gas. A preliminary study was done to select the optimal treatment time to be applied for each essential oil: these samples were referred to as P30LA and P90TT, for plasma treatment time of 30 and 90 seconds. A series of samples of cotton and viscose fabrics were not immersed into the oils, but were subjected to the same plasma treatment conditions, and were designated as P30 and P90. Moreover, fabric samples dipped into lavender and tea tree oil without plasma treatment were also prepared and were notated as P0LA and P0TT. Furthermore, fabrics without any treatment (P0) were also characterized.

Sample characterization

Contact angle measurements and surface energy determination

Contact angle measurements for untreated and treated fabrics were carried out using an apparatus equipped with a video camera and computer software (OCAH 200 DataPhysicsInstruments). The surface of both treated and untreated fabrics were investigated. Distilled water and methylene iodide (Aldrich, 99% purity) were used as probe liquids. A droplet of the probe liquid was deposited on the fabric surface by a micro-syringe. Contact angle values and droplet volumes were recorded for 30 seconds.

The total components of the apparent surface energy (γ), its polar (γ^p) and dispersive (γ^d) contributions were calculated from the contact angle values, according to Owens-Wendt's approach for both fabrics and treatment conditions.³⁸ The contact angle values used in surface energy calculations were those measured after the drop shape reach its equilibrium on the fabric surface. The surface tension of the test probe liquids was reported in the literature.³⁹

ATR-FTIR analysis

The chemical changes occurring during the plasma treatment with or without oil immersion were analyzed using ATR-FTIR. The spectra were recorded using a Thermo Scientific Nicolet iS10 FTIR spectrometer in the range of 500-4000 cm^{-1} with a resolution of 4 cm^{-1} for 64 scans per spectrum.

Carboxyl content

The carboxyl content in all samples was estimated using the sodium bicarbonate/sodium chloride test, according to TAPPI 237 cm - 08.⁴⁰ Briefly, disintegrated samples were extracted with diluted hydrochloric acid, washed, reacted with sodium bicarbonate/sodium chloride solution and then filtered. The filtrate is titrated with 0.01 M hydrochloric acid to methyl red end point. The titration value was recorded and the carboxyl content calculated according to the formula presented in the standard.

Colorimetric test for aldehyde groups

The presence of aldehyde groups on the two fabrics was described using the colorimetric test described in Malek and Holme.²⁹ Aldehyde groups reduce Fehling's solution to give a precipitate of cuprous oxide. The areas on the fibers containing -CHO groups will be colored pink or red and may be seen under the microscope. The test was performed as follows: small pieces of fabric samples were boiled in 10 ml of Fehling's solution for 10 min. The samples were removed and washed with water, while the remaining solution was filtered. Both washed samples and filter paper were examined under an optical microscope to detect red coloration.

Hydroxyl end groups

The hydroxyl end groups in the untreated and oil-treated fabrics were determined after dyeing the samples with reactive Black 5. The dyeing was performed with 2% of dye (relative to the weight of fibers), with a liquor ratio of 1:100, at pH 11 using 20 mg/ml of sodium carbonate and 60 mg/ml of sodium sulphate at 60 °C for 90 min with mechanical agitation of 30 rpm. The samples were all dyed in the same bath at the same time. After that, the samples were washed in hot and cold water and then dried in an oven at 40 °C for 24 h. Afterward, the reflection factor (R) was measured at 590 nm using a Spectraflash SF300. The K/S values were calculated according to Kubelka-Munk's equation, where K and S stand for the absorption and scattering coefficient, respectively.⁴¹ An increase in the K/S value indicates a higher number of hydroxyl groups, which react covalently with this reactive dye.^{42,43}

Qualitative assessment of antimicrobial activity

The antimicrobial activity of the fabrics was assessed using the parallel streak method AATCC-TM 147-2004.⁴⁴ The test organisms used were Gram-negative *Escherichia coli* ATCC 8739 and Gram-positive *Staphylococcus aureus* ATCC 6538. The inoculum was prepared by transferring 1.0 \pm 0.1 mL of a 24 h broth culture of each bacterium into a test tube containing 9.0 \pm 0.1 mL of sterile distilled water, followed by an adequate stirring process. One loop full of the diluted inoculum was loaded and transferred to the surface of the sterile nutrient agar plate by making five parallel inoculum streaks of approximately 60 mm in length, spaced 10 mm apart, covering the central area of the Petri dish without refilling the loop. The test specimen was gently pressed transversely, across the five inoculums of streaks to ensure intimate contact with the agar surface. The plates were incubated at 37 \pm 2 °C for 18-24 h; a clear area of interrupted growth along the sides of the samples indicated the antibacterial activity of the fabrics.

RESULTS AND DISCUSSION**Contact angles and surface energy**

Cellulosic textiles, such as cotton and viscose, present quite different physicochemical properties, as can be seen in Table 1. Viscose fabric has higher apparent surface energy and a lower polar component than cotton fabric. The plasma treatment of the cotton and viscose fabrics under argon atmosphere leads to a decrease of the water contact angle and an increase of the surface energy, as shown in Table 1. Moreover, this trend increases with plasma treatment time. This result was due to the surface oxidation and was also found by Gaiolas *et al.*²⁸ On the other hand, the immersion of the fabrics into lavender and tea tree oils without further plasma treatment affects significantly the physicochemical properties of the fabrics. Lavender essential oil increases the apparent surface energy by the increase of the dispersive component, this effect being higher for cotton fabric. In addition, for the cotton fabric, the polar component was also affected. The immersion into tea tree essential oil also increases the surface energy, modifying the dispersive and polar components. These results are due to the large number of hydrocarbon based components with oxygen containing groups (hydroxyl, ester, ketone and ether groups) present in the oils.^{16,18,20-22,25} Concerning the plasma treatment of the oil immersed fabrics, from preliminary trials for

lavender oil, differences in contact angles for an exposure time of 30 or 90 s were not verified (data not shown). So, the samples immersed into lavender oil and subjected to 30 s of plasma treatment were fully characterized. For the samples grafted with tea tree oil, it was verified that an increase of exposure time to the plasma discharge from 30 to 90 s had a greater impact on contact angles, and thus only the samples treated for 90 s were used for further experiments. An increase of the water contact angle was observed for the oil immersed fabrics treated with plasma, compared to the corresponding oil immersed fabrics without plasma treatment (Table 1). Moreover, the evolution of the water contact angle at the surface of the oil immersed and plasma treated samples shows that it is far more stable compared to that of the other samples, once the liquid penetration was significantly reduced, as can be seen in Figs. 1 and 2, for cotton fabric with lavender oil and viscose fabric with tea tree oil, respectively.

Additionally, the plasma treatment of oil immersed fabrics decreased the surface free energy, excepting the CO-P90TT sample. These results indicated that the resulting polymer is hydrocarbon dense and is unlikely that a reorientation of polar groups occurs at the solid-liquid interface.

Table 1

Water contact angle (θ_w) and apparent surface energy of untreated cotton and viscose fabrics (CO-P0 and CV-P0 respectively), plasma treated (P30; P90), oil immersed (P0LA; P0TT), as well as oil immersed and plasma treated fabrics (P30LA; P90TT)

Sample	θ_w (°)	Surface energy (mN/m)		
		Dispersive	Polar	Total
CO-P0	103.4 ± 6.4	7.5	4.5	12.0
CO-P30	91.6 ± 7.0	33.4	1.4	34.8
CO-P90	93.9 ± 1.1	36.8	0.6	37.4
CO-P0LA	100.0 ± 4.8	40.3	0.0	40.3
CO-P30LA	132.1 ± 0.6	28.8	3.3	32.1
CO-P0TT	120.7 ± 4.7	19.1	0.1	19.2
CO-P90TT	125.4 ± 5.3	32.3	2.5	34.8
CV-P0	96.3 ± 4.3	30.4	0.9	31.3
CV-P30	89.6 ± 1.5	34.4	1.7	36.1
CV-P90	72.2 ± 7.1	31.1	9.1	40.2
CV-P0LA	96.0 ± 4.0	36.1	0.4	36.5
CV-P30LA	120.0 ± 3.7	30.0	1.1	31.1
CV-P0TT	80.6 ± 5.3	44.1	2.5	46.6
CV-P90TT	107.7 ± 6.9	42.2	0.5	42.7

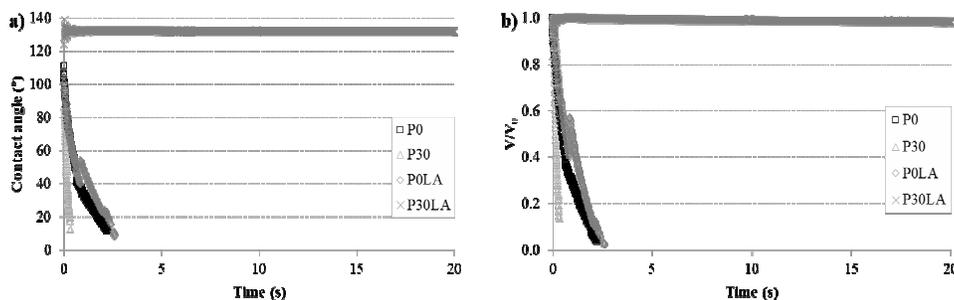


Figure 1: Evolution of water contact angle (a) and water droplet volume (b) at the surface of cotton fabric without any treatment (P0), plasma treated (P30), immersed into lavender essential oil (POLA) and immersed into lavender essential oil with plasma treatment (P30LA)

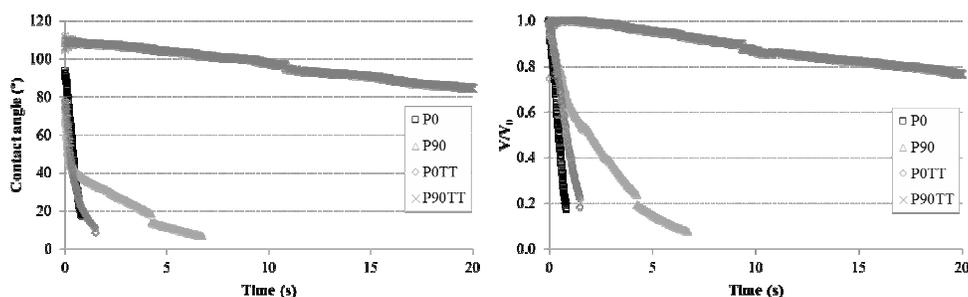


Figure 2: Evolution of water contact angle (a) and water droplet volume (b) at the surface of viscose fabric without any treatment (P0), plasma treated (P90), immersed into tea tree essential oil (P0TT) and immersed into tea tree essential oil with plasma treatment (P90TT)

Thus, the polymer is structurally stable in contact with water and can be a barrier against water in the fabric samples. These outcomes can be attributed to grafting of the different compounds that constitute the oils. As lavender and tea tree oils are a mixture of several components with different vapor pressures, it is not clear which components contribute to the composition of the resulting polymer. However, in the case of lavender oil, linalyl acetate and linalool are the main constituents that can attach to cellulose by hydrogen bonding from the hydroxyl and carboxyl groups. Moreover, interactions between these constituents and cellulose may also occur through dispersive London-type forces. According to Tone *et al.*,³⁷ the activation of the double bonds may take place during plasma polymerization, and thus the polymerization is accelerated. Therefore, the fixation of the compounds is highly determined by their structure and presence of double bonds.³⁷ Easton *et al.*³⁵ found that the polymerisation of the monomers present in the lavender essential oil resulted in a polymer with characteristics that ranged from mildly hydrophilic to mildly

hydrophobic, depending upon the radiofrequency (RF) power used. For the tea tree essential oil, terpinen-4-ol might be the main responsible for the changes in the physicochemical properties of the fabrics, which can bind to cellulose through hydrogen bonds or by dispersive London-type forces. Also, Bakaza *et al.*^{31,32} observed that plasma deposited coatings of terpinen-4-ol change the physicochemical properties of glass substrates and significantly increase water contact angles.

Chemical analysis –ATR-FTIR analysis

The different samples were then characterized by ATR-FTIR spectroscopy. This technique did not afford a clear cut-evidence of the chemical changes achieved, since the FTIR spectra of the untreated and treated fabrics were very similar (Fig.3). This could be attributed to the fact that the depth of analysis in ATR-FTIR is of a few micrometers, too big to give rise to any appreciable detection of the grafted compounds, as they are present in very low amounts, compared to cellulose. However, in the spectrum of CO-P30LA, the appearance of a band at 1732cm^{-1} was observed and assigned to C=O

stretching resulting from carboxyl and aldehyde groups on this sample or from the ester groups present in lavender oil compounds as linalyl acetate.^{11,35} Moreover, the peak around 895 cm^{-1} confirms the presence of the C-O-C glycosidic bond in cotton fabric and the intensity of the peak remained almost unchanged after the different treatments.³⁰ Thus, the appearance of the carbonyl group (1732 cm^{-1}) in the CO-P30LA sample was not due to the cleavage of the $\beta(1-4)$ glucosidic linkages, followed by oxidation. Concerning the viscose fabrics, the presence of carbonyl groups was observed in all spectra (Fig. 3).

To quantify the carboxyl group contents, detect the presence of aldehyde groups and evaluate the hydroxyl end groups, different tests were done and the results are reported in Table 2. As shown in this table, compared to the viscose, the cotton fabric had fewer carboxyl and hydroxyl

groups, and the presence of aldehyde groups was not detected at all. These differences may be explained by the oxidative degradation of cellulose in the viscose production process, where hydroxyl groups are converted to aldehyde and carboxyl groups.^{45,46} The differences in functional groups are responsible for the differences found in apparent surface energy (Table 1) and they are in agreement with those observed in the ATR-FTIR spectra of the cotton and viscose samples (CO-P0 and CV-P0, Fig. 3). It was verified that all the treatments performed on the cotton fabrics increased the carboxyl content, lead to the presence of aldehyde groups and had no effect on hydroxyl end groups. Other studies showed that plasma treatment of cotton resulted in the formation of carboxyl groups, but the concentration of aldehyde groups depended on whether the gas used was air or oxygen.^{12,30}

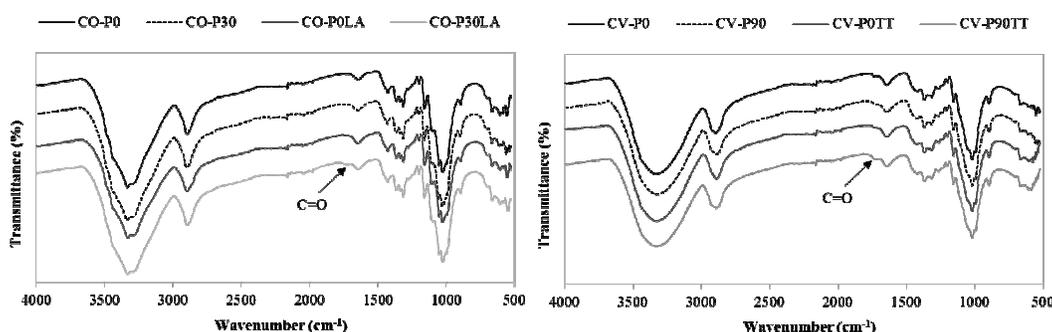


Figure 3: ATR-FTIR spectra of untreated and plasma treated cotton fabric with lavender essential oil and untreated and plasma treated viscose fabric with tea tree essential oil

Table 2
Carboxyl, aldehyde and hydroxyl groups of untreated and treated fabric samples

Sample	Carboxyl group content (meq/100 g)	Aldehyde group inference	K/S value (hydroxyl end groups)
CO-P0	3.3	No red precipitate	2.6
CO-P30	11.4	Red precipitate	2.8
CO-P90	12.7	Red precipitate	2.7
CO-P0LA	12.9	Red precipitate	2.7
CO-P30LA	10.1	Red precipitate	2.8
CO-P0TT	12.2	Red precipitate	2.8
CO-P90TT	12.6	Red precipitate	2.6
CV-P0	9.0	Red precipitate	5.0
CV-P30	13.2	Red precipitate	6.1
CV-P90	10.0	Red precipitate	5.7
CV-P0LA	7.4	Red precipitate	4.6
CV-P30LA	7.9	Red precipitate	4.5
CV-P0TT	13.5	Red precipitate	5.3
CV-P90TT	14.7	Red precipitate	4.6

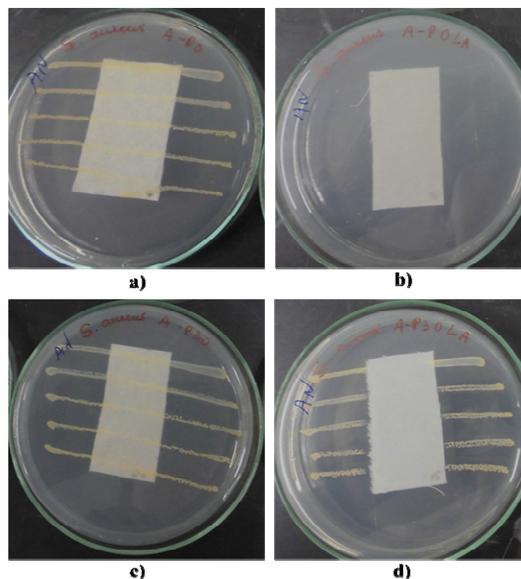


Figure 4: Antimicrobial activity of untreated (a) CO-P0) and treated cotton fabric (b) CO-P0LA, c) CO-P30, d) CO-P30LA) using parallel streak test for *S. aureus*

Concerning the viscose fabrics, all the samples had aldehyde groups, but the treatment with lavender oil or tea tree oil led to different results concerning the carboxyl and hydroxyl end groups. Thus, the samples immersed into lavender oil without and with plasma treatment presented the lowest content of carboxyl and hydroxyl groups, the value being even lower than that for the initial sample (CV-P0). In turn, immersion into tea tree oil led to an increase of carboxyl groups. This result is in agreement with the findings of Bakaza *et al.*,^{31,32} who observed that plasma polymerisation of terpinen-4-ol led to the formation of carboxylic acid groups.

Antimicrobial activity

In the present study, the antimicrobial activity of the cotton and viscose fabrics treated with both oils and subjected or not to plasma treatment against *S. aureus* and *E. coli* bacteria were evaluated by visual inspection of the plates, after incubation. The interrupted growth underneath and along the sides of the test material indicates the antimicrobial activity of the sample. The growth inhibition was only observed for the cotton fabric immersed into lavender oil, without plasma treatment (CO-P0LA) against *S. aureus* (illustrated in Fig. 4). It was not possible to measure the zone of inhibition, since no growth was detected. None of the samples exhibited antimicrobial activity against *E. coli*. The differences in antibacterial efficiency between the

bacteria may reflect differences in their cell wall structure, and the susceptibility to destruction and disruption.^{47,48} Better antibacterial activity of nanoparticles, compounds or essential oils against Gram-positive compared to Gram-negative bacteria was also found in other studies.^{9,16,22,36,47,48}

According to the literature, the minimum inhibitory concentration (MIC) values for lavender oil is in general higher against *E. coli* than against *S. aureus*.^{16,21,22} Thus, the amount of lavender essential oil present in the analysed fabrics may not reach the MIC value for *E. coli*. On the other hand, the MIC values are higher for lavender oil than for tea tree oil,^{21,24} and thus growth inhibition would be expected for the samples treated with tea tree oil. However, de Rapper *et al.*¹⁷ confirmed a higher MIC value for tea tree oil, compared to lavender oil, against *S. aureus*. These discrepant results might be explained by the amount of active compounds present in the commercial oils. According to Danh *et al.*¹⁶, the extraction methods of the lavender essential oil influence its antimicrobial activity, since they allow extracting different amounts of active compounds. The differences in the antimicrobial activity of the cotton and viscose fabrics immersed into lavender oil (CO-P0LA and CV-P0LA) may arise from the functional groups present in these fabrics, which may interact with the essential oil differently, or simply due to the

more open structure of the viscose fabric, which can entrap a lower amount of oil.

Concerning the plasma treatment of the oil immersed fabrics, it was observed that none of the samples exhibited antimicrobial activity after the treatment. Moreover, comparing the cotton sample immersed into lavender oil without plasma treatment (CO-P0LA) with the plasma treated one (CO-P30LA), it can be seen that the plasma treatment had a detrimental effect. This may reflect the variation in the chemical and physicochemical properties between the two samples (as shown in Tables 1 and 2), the ability to bind or entrap the essential oil, the extent of oil release from the sample and diffusion into agar. Furthermore, the polymer structure resulting from plasma polymerization may possess different chemical and physical properties from those of its precursors, and this alters the extent of the antimicrobial activity, as shown by Bakaza *et al.*³¹ According to these authors, the plasma polymerization of terpinen-4-ol at a lower input RF power preserves the original structure of this monomer responsible for the antibacterial inhibitory properties, contrarily to the use of a higher RF power. Thus, the results suggest that the plasma conditions used in the present study were not adequate to preserve the chemical structure of the compounds accountable for the antimicrobial activity of the lavender oil.

CONCLUSION

A plasma polymerization route was used to modify cotton and viscose fabrics by application of lavender and tea tree essential oils. Compared to control fabrics (CO-P0 and CV-P0), plasma treatment, immersion into essential oils and plasma polymerization of the compounds present in essential oils alter the physicochemical properties and the chemical structure of the fabrics. In fact, the compounds existing in essential oils were chemically grafted to cotton and viscose fabrics, rendering them more stable in contact with water, since the water contact angle increased and the water penetration into the fabrics was reduced. All the treatments led to an increase of the carboxyl group contents, except for the viscose sample treated with lavender oil both without and with plasma treatment (CV-P0LA; CV-P30LA). These last samples had a lower amount of hydroxyl end groups compared to the control viscose fabric, while all the treatments performed on the cotton fabric had almost no effect on the hydroxyl end groups.

None of the samples exhibited antimicrobial activity against *E. coli*, while the cotton fabric immersed into lavender oil (CO-P0LA) showed antibacterial inhibitory properties against *S. aureus*. However, the polymerization of the compounds present in lavender oil onto cotton fabric by plasma (sample CO-P30LA) resulted in the loss of the inhibitory effect, probably due to the fragmentation of the compounds responsible for antimicrobial activity. The application of a solvent free process, such as cold plasma treatment, and the use of a product derived from renewable sources as the essential oils, may constitute an innovative approach to functional finishes of textile fabrics. However, in order to preserve the antimicrobial activity of essential oils, it is necessary to optimize the plasma treatment conditions.

REFERENCES

- ¹ A. Mariscal, R. M. Lopez-Gigosos, M. Carnero-Varo and J. Fernandez-Crehuet, *Eur. J. Clin. Microbiol. Infect. Dis.*, **30**, 227 (2011).
- ² S. Shahidi, N. Aslan, M. Ghoranneviss and M. Korachi, *Cellulose*, **21**, 1933 (2014).
- ³ Š. Olivera and J. Volmajer-Valh, *Fibres Text. East. Eur.*, **5**, 24 (2013).
- ⁴ C. W. Kan, Y. L. Lam, C. W. M. Yuen, A. Luximon, K. W. Lau *et al.*, *J. Phys.: Conf. Ser.*, **441**, 1 (2013).
- ⁵ A. Kramar, V. Prysiaznyi, B. Dojčinović, K. Mihajlovsk, B. M. Obradović *et al.*, *Surf. Coat. Tech.*, **234**, 92 (2013).
- ⁶ Y. L. Lam, C. W. Kan and C. W. M. Yuen, *Fiber. Polym.*, **14**, 52 (2013).
- ⁷ Z. Peršin, M. Devetak, I. Drevenšek-Olenik, A. Vesel, M. Mozetič *et al.*, *Carbohydr. Polym.*, **97**, 143 (2013).
- ⁸ G. Thilagavathi, K. Rajendrakumar and R. Rajendran, *Colourage*, **53**, 49 (2006).
- ⁹ N. A. Ibrahim, B. M. Eid, M. M. Hashem and R. Refai, *J. Ind. Text.*, **39**, 233 (2010).
- ¹⁰ E. Nithya, R. Radhai, R. Rajendran, S. Jayakumar and K. Vaideki, *Carbohydr. Polym.*, **88**, 986 (2012).
- ¹¹ S. Nourbakhsh and A. Ashjarian, *Materials*, **5**, 1247 (2012).
- ¹² K. Vaideki, S. Jayakumar and R. Rajendran, *Plasma Chem. Plasma P.*, **29**, 515 (2009).
- ¹³ M. D. Balakumaran, R. Ramachandran, S. Jagadeeswari and P. T. Kalaichelvan, *Int. Biodeter. Biodegr.*, **107**, 48 (2016).
- ¹⁴ I. H. N. Bassolé and H. R. Juliani, *Molecules*, **17**, 3989 (2012).
- ¹⁵ C. F. Carson, K. A. Hammer and T. V. Riley, *Clin. Microbiol. Rev.*, **19**, 50 (2006).

- ¹⁶ L. T. Danh, L. N. Han, N. D. A. Triet, J. Zhao, R. Mammucari *et al.*, *Food Bioprocess Tech.*, **6**, 3481 (2013).
- ¹⁷ S. de Rapper, G. Kamatou, A. Viljoen and S. van Vuuren, *Evid.-Based Compl. Alt.*, **2013**, 1 (2013).
- ¹⁸ K. A. Hammer, C. F. Carson and T. V. Riley, *J. Appl. Microbiol.*, **95**, 853 (2003).
- ¹⁹ C. Jianu, G. Pop, A. T. Gruia and F. G. Horhat, *Int. J. Agric. Biol.*, **15**, 772 (2013).
- ²⁰ J. May, C. H. Chan, A. King, L. Williams and G. L. French, *J. Antimicrob. Chemother.*, **45**, 639 (2000).
- ²¹ L. Mayaud, A. Carricajo, A. Zhiriand G. Aubert, *Lett. Appl. Microbiol.*, **47**, 167 (2008).
- ²² M. Soković, J. Glamočlija, P. D. Marin, D. Brkić and L. J. L. D. van Griensven, *Molecules*, **15**, 7532 (2010).
- ²³ L. Stanojević, M. Stanković, M. Cakić, V. Nikolić, L. Nikolić *et al.*, *Hem. Ind.*, **65**, 455 (2011).
- ²⁴ N. Thosar, S. Basak, R. N. Bahadure and M. Rajurkar, *Eur. J. Dent.*, **7**, 71 (2013).
- ²⁵ S. F. Van Vuuren, G. P. P. Kamatou and A. M. Viljoen, *S. Afr. J. Bot.*, **76**, 686 (2010).
- ²⁶ N. Abidi and E. Hequet, *J. Appl. Polym. Sci.*, **93**, 145 (2004).
- ²⁷ N. V. Bhat, A. N. Netravali, A. V. Gore, M. P. Sathianarayanan, G. A. Arolkar *et al.*, *Text. Res. J.*, **81**, 1014 (2011).
- ²⁸ C. Gaiolas, M. N. Belgacem, L. Silva, W. Thielemans, A. P. Costa *et al.*, *J. Colloid Interf. Sci.*, **330**, 298 (2009).
- ²⁹ R. M. A. MalekandI. Holme, *Iran. Polym. J.*, **12**, 271 (2003).
- ³⁰ E. Nithya, R. Radhai, R. Rajendran, S. Shalini, V. Rajendran *et al.*, *Carbohydr. Polym.*, **83**, 1652 (2011).
- ³¹ K. Bakaza, M. V. Jacob, V. K. Truong, F. Wang, W. A. A. Pushpamali *et al.*, *Biomacromolecules*, **11**, 2016 (2010).
- ³² K. Bakaza, M. V. Jacob, R. J. Crawford and E. P. Ivanova, in *Procs. ICAR2010*, Valladolid, November 3-5, 2011, pp. 175-179.
- ³³ K. Bakaza, M. V. Jacob, R. J. Crawford and E. P. Ivanova, *Acta Biomater.*, **7**, 2015 (2011).
- ³⁴ C. D. Easton and M. B. Jacob, *Polym. Degrad. Stabil.*, **94**, 597 (2009).
- ³⁵ C. D. Easton, M. B. Jacob and R. A. Shanks, *Polymer*, **50**, 3465 (2009).
- ³⁶ A. Pegalajar-Jurado, C. D. Easton, K. E. Styan and S. L. McArthur, *J. Mater. Chem. B*, **2**, 4993 (2014).
- ³⁷ S. Tone, T. Masawaki and K. Eguchi, *J. Membrane Sci.*, **118**, 31 (1996).
- ³⁸ D.K. Owens and R.C. Wendt, *J. Appl. Polym.Sci.*, **13**, 1741 (1969).
- ³⁹ R. J. GoodandC. J. van Oss, in “Modern Approaches to Wettability: Theory and Applications”, edited by M. E. Schrader and G. I. Loeb, Plenum Press, 1991, New York, pp. 1-27.
- ⁴⁰ TAPPI 237 cm-08 in “TAPPI Test Methods”, TAPPI Press, Atlanta, 2008.
- ⁴¹ P. Kubelka and F. Munk, *Z. Tech. Physik*, **12**, 593 (1931).
- ⁴² I. C. Gouveia, C. A. Laura and A. P. Gomes, *J. Text. I.*, **102**, 203 (2011).
- ⁴³ A. O’Neill and A. Cavaco-Paulo, *Biocatal. Biotransform.*, **22**, 353 (2004).
- ⁴⁴ AATCC-TM 147-2004 in “AATCC Technical Manual”, American Association of Textile Chemists and Colorists, Research Triangle Park, U.S.A., 2005.
- ⁴⁵ R. R. Mather and R. H. Wardman, in “The Chemistry of Textile Fibres”, The Royal Society of Chemistry, 2015, pp. 111-143.
- ⁴⁶ A. Potthast, J. Röhrling, T. Rosenau, A. Borgards, H. Sixta *et al.*, *Biomacromolecules*, **2003**, 743 (2003).
- ⁴⁷ G. Fu, P. S. Vary and C.-T. Lin, *J. Phys. Chem. B*, **109**, 8889 (2005).
- ⁴⁸ K. Page, R. G. Palgrave, I. P. Parkin, M. Wilson, S. L. P. Savin *et al.*, *J. Mater. Chem.*, **17**, 95 (2007).