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CHARACTERIZATION OF TEA AQUEOUS EXTRACTS AND THEIR UTILIZATION FOR DYEING AND FUNCTIONALIZING FABRICS OF DIFFERENT CHEMICAL COMPOSITIONS

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Green, black, rooibos, and hibiscus tea (GT, BT, RT, and HT) aqueous extracts were prepared and characterized in terms of total flavonoids (*TFC*) and total phenolic (*TPC*) contents and antioxidant and antimicrobial activities. BT has the highest, while HT has the lowest *TFC* (1213 *vs*. 415 mg l^{-1}), while the extracts' *TPC*s (2283 – 7251 mg l^{-1}) decreased in the following order: BT > GT > RT > HT. Their antioxidant activities of 78.1 – 93.1% and 97.8 – 100% were determined according to DPPH and ABTS methods, respectively. BT and especially GT aqueous extracts possessed mild effects against several microorganisms. All examined extracts have an affinity for dyeing wool, cellulose acetate, polyamide, and cotton, which is proven by the color strength values of 1.65–19.12. Wool, polyacrylonitrile, polyester, polyamide, cotton, and cellulose acetate functionalized with GT aqueous extract inhibited the growth of *S. aureus* and *E. coli*, while polyacrylonitrile and cotton also inhibited the growth of *E. faecalis*, and *C. albicans*, respectively. Wide inhibition zones for *S. aureus* were observed for fabrics functionalized with BT aqueous extract. Generally, the investigated fabrics showed very high (81.60 – 100%) ABTS radical scavenging ability independent of the extract used. *TPC*s have good linear correlations with the antioxidant activities of wool and polyacrylonitrile determined by the DPPH method. Fabrics with different chemical compositions dyed and/or functionalized with GT or BT aqueous extracts can be used to produce high-value-added medical textiles with therapeutic, prophylactic, and protective functions. They can find potential applications in wound treatment, especially in skin wounds that are susceptible to infection with *S. aureus*. Moreover, wool and cotton functionalized with GT or BT aqueous extract can also be considered for use in disposable medical textiles like bandages and gauze used in the wound-healing process.

Keywords: tea aqueous extract; fabric; functionalization; dyeing; antioxidant and antimicrobial activity

КАРАКТЕРИЗАЦИЈА НА ВОДНИ ЕКСТРАКТИ ОД ЧАЕВИ И НИВНА УПОТРЕБА ЗА БОЕЊЕ И ФУНКЦИОНАЛИЗАЦИЈА НА ТКАЕНИНИ СО РАЗЛИЧЕН ХЕМИСКИ СОСТАВ

Водните екстракти подготвени од црн, зелен, ројбуш и хибискус чај (GT, BT, RT и HT) се карактеризирани од аспект на количество вкупни флавоноиди (*TFC*) и вкупни феноли (*TPC*), и антиоксидациска и антимикробна активност. BT покажува најголема, а HT најмала *TFC* (1213 *vs*. 415 mg l^{-1}), додека *TPC* во воденте екстракти (2283 – 7251 mg l^{-1}) се намалува според следниов редослед: BT > GT > RT > HT. Нивната антиоксидациска активност, одредена користејќи ги методите DPPH и ABTS, се движи соодветно во опсег 78,1 – 93.1 % и 97,8 – 100 %. Водните екстракти на BT и GT покажаа благ ефект врз одредени микроорганизми. Сите испитувани екстракти имаат афинитет за боење волна, целулозен ацетат, полиамид и памук, што е потврдено со одредување на јачина на бојата на ткаенините која изнесува 1,65 – 19.12. Волната, полиакрилонитрилот, полиестерот, полиамидот, памукот и целулозниот ацетат функционализирани со воден екстракт од GT го инхибираат растот на *S. aureus* и *E. coli*, додека полиакрилонитрилот и памукот соодветно го инхибираат растот на *E. faecalis* и *C. albicans*. Широки инхибициони зони за *S. aureus* се воочени за ткаенините функционализирани со BT. Општо, испитуваните ткаенини покажуваат многу висока (81,60 – 100 %) способност за отстранување на ABTS-радикали. Утврдена е добра линеарна зависност помеѓу *TPC* во водните екстракти и антиоксидациската активност на волната и полиакрилонитрилот одредени преку методот DPPH. Ткаенините со различен хемиски состав обоени и/или функционализирани со водни екстракти од GT или BT можат да се користат за производство на медицински текстил со висока додадена вредност, кој има терапевтски, профилактички и заштитни функции. Медицинските текстилни материјали имаат потенцијална примена за третман на рани, особено за рани на кожа кои се подложни на инфекција со *S. aureus*. Покрај тоа, волната и памукот функционализирани со водни екстракти од GT или BT можат да се користат за производство на медицински завои и гази за еднократна употреба.

Клучни зборови: воден екстракт од чај; ткаенина; функционализација; боење; антиоксидациска и антимикробна активност

1. INTRODUCTION

Within the era of fast fashion, Western clothing brands originating from wealthy countries have outsourced the fiber, yarn, and fabric dyeing processes to developing countries (like Bangladesh, China, Thailand, and Indonesia) where textile workers are exposed to harmful chemicals. Conventional textile dyeing and finishing processes can require as much as 200 tons of water for every ton of produced textiles, and the majority of this water, laden with residual dyes, hazardous chemicals, heavy metals, microfibers, and mordants, returns to nature, which is still one of the main environmental issues.[1] Moreover, textile dyeing and finishing are responsible for 3 % of global CO_2 emissions.[2] At the end of the cycle, the garments dyed in developing countries are sent back to consumers in wealthy countries. To break the above-discussed cycle of fast fashion, consumers should choose quality over quantity, respect each garment and its creators, and opt for a minimal negative impact on people, animals, and the planet. In light of that, the non-toxic and biodegradable dyes and colorants from sustainable sources like plants,[3–6] wood-based materials,[7] fungi[8], and bacteria[9] having no or minimal environmental impact have been within the scope of many researchers. Although the application of some of the mentioned natural dyes dates back to pre-historic times, only 10 % of today's textiles are dyed with natural dyes.[10]

Since sustainable fashion brands favor natural dyes and multifunctional clothes more progressively, this paper focuses on a green, clean, chemical-free, and sustainable method for fabric dyeing and/or functionalization. In order to obtain a palette of fabrics having different colors and shades, intensely and differently colored aqueous extracts of green, black, rooibos, and hibiscus teas were employed as dyeing agents for wool, cotton, cellulose acetate, polyacrylonitrile, polyester, and polyamide fibers. These herbal teas were not chosen randomly. Green and black teas were selected due to their abundance of polyphenolic compounds, mainly phenolic acids and flavonoids that are known for their antioxidant and various biological activities, such as anticancer, anti-inflammatory, antitumor, antibacterial, and antiviral activities.[11] Moreover, the literature data regarding the characterization and utilization of rooibos and hibiscus teas as natural colorants and resources of bioactive compounds that can functionalize and/or dye fabrics of different compositions were very rare and not detailed enough. The results obtained for the total flavonoids and phenolic contents, as well as antioxidant and antimicrobial activities of examined tea aqueous extracts, indicated that they offer excellent potential for developing bioactive clothing. The demand for bioactive textiles, especially textiles with strong antimicrobial activity, rapidly increased during the COVID-19 pandemic. In that context, antimicrobial and antioxidative activities of all fabrics were tested as essential properties responsible for microbial inhibition and the reduction of inflammation, respectively. At the end of this investigation and depending on the different manifestations of antioxidant and antimicrobial activities, the potential applications of functionalized fabrics were proposed.

2. EXPERIMENTAL

2.1. *Preparation of tea aqueous extracts*

The commercially available (bought from Tea House, Belgrade, Serbia) dried green (*Camel-*

lia sinensis originating from Sri Lanka), black (*Camellia sinensis* originating from Sri Lanka), rooibos (*Aspalathus linearis* grows in the vicinity of the Cederberg Mountains in South Africa), and hibiscus (*Hibisci flos* originating from Sudan) teas having 4.71, 4.14, 6.91, and 5.75 % (m/m) moisture were firstly ground into a fine powder using a mill. Their extracts were prepared by immersing 20 g of each tea in 1 l of distilled water. The temperature of the mixtures was raised to the boiling point and kept at this temperature for 5 min. Thereafter, the mixtures were allowed to cool for 2 h, and the solid parts were separated by filtration using qualitative filter paper (73 g/m^2) having a pore size of 5 to 13 μm supplied by Lab Logistics Group GmbH. Extracts were prepared following the procedure published by Čuk et al.[12] The extracts were immediately characterized and used for fabric dyeing and functionalization. In further text, the following abbreviations are used for green, black, rooibos, and hibiscus teas: GT, BT, RT, and HT, respectively.

2.2. *Characterization of the prepared extracts*

All spectrophotometric measurements were performed on a Beckman DU-650 spectrophotometer. The total flavonoid and phenolic contents (*TFC* and *TPC*) within the tea extracts were determined spectrophotometrically according to the methods described by Pavun *et al*.[13] In the case of determination of *TFC*, quercetin was used for the construction of a calibration curve (standard solutions 12.5–100 mg ml^{-1} in 80 % (v/v) ethanol). 0.5 ml of the standard quercetin solutions or tea aqueous extracts were mixed with 1.5 ml of 95 % (v/v) ethanol, 0.1 ml of 10 % (m/v) $Al(NO_3)_3$, 0.1 ml of 1 mol l^{-1} CH_3CO_2K, and 2.8 ml of deionized water. In the blank, the volume of 10 % (m/v) $Al(NO_3)_3$ was substituted by the same volume of deionized water. After incubation at room temperature for 30 min, the reaction mixtures' absorbances were measured at λ = 415 nm. The Folin-Ciocalteu (FC) method was used to determine *TPC* in the tea aqueous extracts. Briefly, 1 ml of the prepared extract was added into a 25 ml volumetric flask containing 9 ml of water and 1 ml of FC reagent. After 5 min, 10 ml of Na_2CO_3 solution was added, and the remaining volume of the flask was filled with deionized water. The reaction took place for 90 min, and then, the solution absorbance was measured at λ = 765 nm against a blank sample consisting of 10 ml of deionized water instead of the extract. The measurements were compared to a standard curve of prepared gallic acid solutions (25–500 mg l^{-1}).

The extract antioxidant activity was evaluated using the DPPH and ABTS assays. According to the first method,[14] 1 ml of each tea aqueous extract was added to 4 ml of a 1 × 10^{-4} mol l^{-1} methanol solution of DPPH. After 60 min of shaking in the dark, the absorbance of the solution was recorded at λ = 517 nm. According to the ABTS method previously described in the literature[15], ABTS$^{\bullet+}$ radical cation solution was prepared by mixing 4.912 ml of ABTS (7 × 10^{-3} mol l^{-1} in phosphate-buffered saline (PBS)) and 0.088 ml of potassium persulfate (0.140 mol l^{-1} in distilled water) solutions. The mixture was stored in the dark and the components were allowed to react for 16 h. Then, the solution was diluted with methanol to obtain an absorbance of 0.700 ± 0 .02 at λ = 734 nm. Two milliliters of this freshly prepared ABTS$^{\bullet+}$ solution was further mixed with 20 µl of each tea aqueous extract, shaken, and stored in the dark for 10 min. After that, the absorbance was measured at the mentioned wavenumber. Inhibition of free radicals by DPPH or ABTS was calculated using Eq. 1:

$$I\ (\%) = \frac{Ac - As}{Ac} \times 100 \qquad (1)$$

where A_c is the absorbance of the control mixture (containing all reagents except the tea aqueous extract), while A_s is the absorbance of the prepared sample containing tea aqueous extract. The determination of *TFC* and *TPC*, as well as extract antioxidant activity was done in triplicate, wherein the coefficients of variation were up to 2.39 %.

The antimicrobial activity of the freshly prepared tea aqueous extracts was studied by the broth microdilution method according to the literature.[16] Seven different laboratory control bacteria strains, *i.e.*, the Gram-positive *Staphylococcus aureus* (ATCC 6538), *Enterococcus faecalis* (ATCC 29212), and *Bacillus subtilis* (ATCC 6633); the Gram-negative *Escherichia coli* (ATCC 8739), *Klebsiella pneumoniae* (NCIMB 9111), *Salmonella typhimurium* (ATCC 14028), and *Pseudomonas aeruginosa* (ATCC 9027), and one yeast, *Candida albicans* (ATCC 10231), were included in the study. The method was performed with an inoculum of microorganisms of approximately 10^6 CFU ml^{-1}, using the MH broth for bacterial strains and Sabouraud dextrose broth (HiMedia Laboratories Pvt. Ltd., Mumbai, India) for *Candida albicans*. To determine the minimum inhibitory concentration (MIC) of the tested samples, the 0.5 ml aliquots of each filtrate were diluted in Mueller Hinton (HiMedia Laboratories Pvt. Ltd., Mumbai, India) broth to the highest concentration. Twofold serial concentrations of the tested samples were prepared in a 96-well microtiter plate (31.2 to 1000 μg ml^{-1}). To make it easier to estimate the effects of the tested tea aqueous extracts, before the incubation step, triphenyl tetrazolium chloride solution (Sigma-Aldrich Company, USA) was added in the final concentration of 0.05 % as a microbial cell growth indicator. The MIC determinations were performed in duplicate, and two positive growth controls were included. Each test was repeated three times, and the mean values are presented.

2.3. *Utilization of extracts for dyeing and functionalizing fabrics*

The Multifiber Adjacent Fabric Style 42 (James Heal, England) was used to evaluate the ability of the prepared extracts to dye worsted wool (WO), polyacrylonitrile (PAN), polyester (PES), polyamide (PA), cotton (CO), and cellulose acetate (CA). Three grams of multifiber fabric (12 cm × 10 cm) was functionalized with 150 ml of extract for 21 h with constant shaking on a water bath (WNE 14, Memmert) at 25 °C. Afterward, the fabrics were rinsed with warm distilled water to remove the unbound dye, dried at room temperature for 48 h, and characterized.

2.4. *Characterization of functionalized fabrics*

The Kubelka-Munk equation[17] was used to calculate the fabrics' color strength (*K/S*) values based on the reflectance recorded on the UV-Vis spectrophotometer (Shimadzu UV-Vis 2600). Fabric reflectance values used for calculating *K/S* were chosen at the same wavenumber that the prepared tea aqueous extract showed the absorption maximum in its UV-Vis spectrum. An example is given in Supplementary Material, Figures S1 and S2.

The fabrics' antioxidant activity was determined according to the DPPH and ABTS methods. The ABTS assay was performed following the method described by Glaser *et al*.[18] One-tenth of a gram of each component of multifiber fabric was added to a test tube containing 3.9 ml of freshly prepared ABTS radical in PBS, and the reaction took place in the dark at 25 °C for 30 min. The radical scavenging activity was evaluated using the absorbance of the solutions at 734 nm and calculated using Eq. 1. The DPPH assay was performed according to the procedure described by Hong,[19] wherein 0.5 g of the multifiber fabric was added to 30 ml of a freshly prepared DPPH methanol solution (1.5×10^{-4} mol l^{-1}). The mixture was kept in the dark for 1 h. After that, the absorbance was measured at λ = 517 nm, while the radical scavenging activity was calculated using Eq. 1. The presented results are the average of three parallel measurements, and the coefficients of variation are below 3.13 %.

The fabrics' antimicrobial activities were tested using the agar-diffusion method according to the previously outlined method[20] with some modifications. The method is based on placing the autoclaved (at 121 °C under a pressure of 1.2 bar for 30 min) fabrics (10 mm × 10 mm) on previously inoculated agar (tryptone soy agar + 0.6% yeast extract) with a bacterial suspension. The widths of the inhibition zones were established after incubation of Petri dishes at 37 °C for 24 h. All experiments were done in triplicate, and the results are presented as the mean value.[21] The positive growth control, without applying the fabrics, was included for each of the tested microorganisms.

3. RESULTS AND DISCUSSION

3.1. *Characterization of tea aqueous extracts*

Before the utilization for fabric dyeing and functionalization, the prepared tea aqueous extracts were characterized in terms of their total flavonoid (*TFC*) and phenolic (*TPC*) contents, antioxidant activity (using DPPH and ABTS assay), and antimicrobial activity.

Among all analyzed samples, BT has the highest *TFC* (1213 mg l^{-1}), while HT has the lowest *TFC* (415 mg l^{-1}), Figure 1. Kaur *et al*.[22] also reported almost three times lower *TFC* in HT than in BT aqueous extract. Furthermore, the results of the current study are also comparable with those published by Aboagye *et al*.[23], who found that BT aqueous extracts contain significantly higher *TFC* than GT aqueous extracts.

From the results (Fig. 1), it is evident that *TPC*, expressed as mg per liter tea aqueous extract, showed considerable variation (2283–7251 mg l^{-1}) and decreased in the following order: BT > GT > RT > HT. These data are in line with those already published. Namely, research reported in papers[24,25] indicated that BT aqueous extract is more polyphenol-rich than the HT aqueous extract. It has to be emphasized that *TPC*s of BT and HT aqueous extracts reported by Büyükbalci and El[24] are significantly lower (1430 and 170 mg l^{-1}, respectively) than those in the current study (7251 and 2282 mg·l^{-1}, respectively), which could be explained by the different experimental conditions used for the tea preparation. Moreover, Samadi and Fard[26] concluded that both the *TFC* and *TPC* of aqueous HT extract were lower than those determined in GT and BT aqueous extracts. Interestingly, the literature survey pointed out that the *TPC* and *TFC* of BT, GT, and RT aqueous extracts were compared in only one paper.[27] According to the results, GT and BT aqueous extracts showed about two times higher *TPC*s than RT tea aqueous extract. GT and BT aqueous extracts examined in the current paper have about 2.2 and 2.8 times higher *TPC*s than RT, Figure 1.

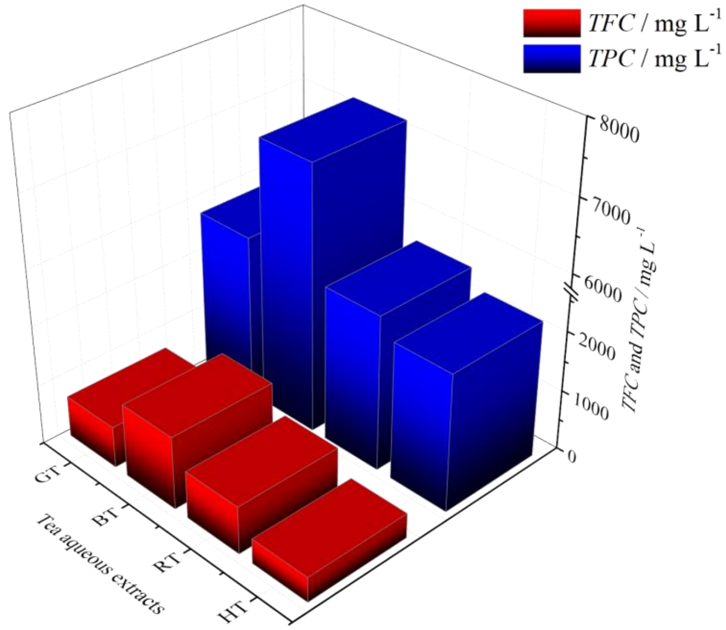

Fig 1. Total flavonoid and phenolic contents (*TFC* and *TPC*, respectively) in the tea aqueous extracts

It is well known that the phenolic content of plant extracts usually correlates with their antioxidant activities.[28] Independent of the applied method (DPPH or ABTS), Oh *et al*.[27] highlighted that among GT, BT, and RT aqueous extracts, the lowest antioxidant activity was observed for RT, which is a direct consequence of its lowest *TPC*. The last one was also evident for the RT aqueous extract prepared in our study, Figures 1 and 2. According to the DPPH method, the calculated RT aqueous extract's antioxidant activity was 78.1%, while GT and BT aqueous extracts possessed antioxidant activities of 92.8 % and 93.1 %, respectively. Although Nadiah *et al*.[29] prepared GT and BT aqueous extracts with 2.5 times higher extract concentration, their antioxidant activities (DPPH method) were significantly lower (21.2 % and 18.8 %, respectively), which could be ascribed to the four times lower temperature (100 *vs*. 25 °C) used for the extracts' preparation. Another confirmation that experimental conditions play an important role in the extract's antioxidant activity lies in the paper[30]. Precisely, low extract concentration (11.8 mg l^{-1}) and applied ultrasound-assisted extraction for 15 min at 55 °C resulted in moderate (68.2 %) to low (46.6 %) antioxidant activity of GT and BT aqueous extracts, respectively.

The tea aqueous extracts' antioxidant activities determined according to the DPPH method differ from those obtained based on the ABTS method (78.1 – 93.1% *vs*. 97.8 – 100%), Fig. 2. The differences in the antioxidant activity assays are expected and can be described by the different reaction mechanisms and the use of different model radicals.[31] Independent of the above discussion, the results obtained in two separate investigations reported almost the same trend, *i.e.*, among GT, BT, and RT aqueous extracts, RT is characterized by the lowest antioxidant activity.

In the next step of this investigation, the antimicrobial activity of tea aqueous extracts was studied based on a determination of MIC, Table 1. The listed results revealed that HT and RT aqueous extracts did not exhibit any antimicrobial effect under the conditions of the performed study. In contrast, BT and especially GT aqueous extracts possessed some mild effects against several microorganisms. Namely, the established MIC values were 500 μg ml^{-1} against *S. aureus* and *E. faecalis* for both BT and GT aqueous extracts, while GT aqueous extract additionally exhibited the same activity against *E. coli* and *C. albicans*. The susceptibility of *S. aureus* to antimicrobial agents can be attributed to the absence of an outer membrane surrounding the cell wall, allowing the diffusion of hydrophobic compounds to the cell interior.[32] The antibacterial effect of GT aqueous extract is mainly due to the presence of catechins which are capable of damaging the cell membrane, inhibiting fatty acid synthesis, and inhibiting enzyme activity.[33] The results obtained in this study are in agreement with the data reported in the literature.[34,35] According to Radji *et al*. [34], the MIC value of GT aqueous extract against *S. aureus* was 400 μg ml^{-1}.

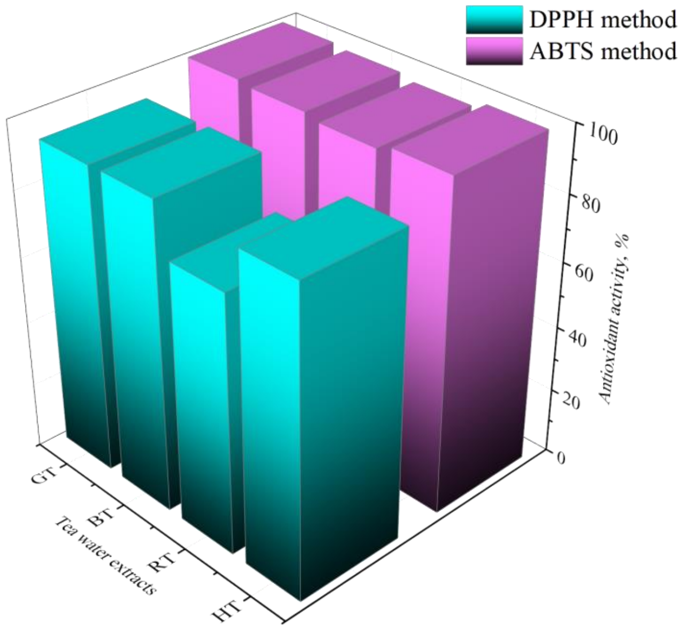

Fig 2. The antioxidant activity of studied tea aqueous extracts determined according to different methods

Table 1

MIC values (μg ml^{-1})

Microorganisms	Tea aqueous extracts			
	GT	BT	RT	HT
S. aureus	500	500	>1000	>1000
E. faecalis	500	500	>1000	>1000
B. subtilis	1000	1000	1000	1000
E. coli	500	>1000	>1000	>1000
K. pneumoniae	>1000	>1000	>1000	>1000
S. typhimurium	>1000	>1000	>1000	>1000
P. aeruginosa	>1000	>1000	>1000	>1000
C. albicans	500	1000	1000	>1000

Taking into account the extracts' abundance of polyphenolic compounds, their excellent antioxidant activity, as well as confirmed microorganisms' sensitivity to BT and especially GT aqueous extracts, the extracts' potential for simultaneous fabric dyeing and functionalization was tested.

3.2. *Dyeing of multifiber fabric using different tea aqueous extracts*

The effectiveness of dyeing different components of multifiber fabric (WO, PAN, PES, PA, CO, and CA) with already characterized tea aqueous extracts has been objectively studied by the fabric color strength (*K/S)* values. From the appearance of multifiber fabrics (Fig. 3) and their *K/S* values (Table 2), it is evident that all tea aqueous extracts are able to dye natural fibers WO, CO, CA, and synthetic fiber PA. Independent of the extract used, *K/S* values decrease in the following order: WO (16.86 – 19.12) > CA (9.39 – 12.96) > PA (7.71 – 11.38) > CO (1.65 – 8.42), Table 2. The observed different shades and color strength values are attributed to the fabrics' different chemical compositions and extracts' nature that dictate the possibility of diverse binding interactions between the colored bioactive compounds' functional groups and fabric surface groups.[36]

WO, PA, CO, and CA functionalized with GT have *K/S* values of 16.86, 7.71, 4.52, and 12.23, respectively (Table 2). These values were calculated based on the corresponding fabrics' reflection at 272 nm (Supplementary material, Fig. S2a). WO and CO have more intensive coloration than PA and CA (Fig. 3), which is ascribed to the fact that employed extracts are mixtures of different compounds. Therefore, the extract adsorption maximum is due to both non-colored and colored extract compounds. On the other hand, the fabric reflection at a certain wavenumber comes from the fabric itself and both colored and non-colored extract compounds competing for a position on its surface. This is the reason behind the inconsistencies between the *K/S* values of fabrics functionalized with GT and RT and their appearances. From the reflection spectra of fabrics functionalized with RT (Supplementary material, Fig. S2b), it seems that in the case of PA, a higher amount of non-colored bioactive compounds absorbing at 283 nm (Supplementary material, Fig. S1) was bound to the fabric surface, contributing to a lower reflection and higher *K/S* value than WO.

Fig 3. The appearance of the dyed multifiber fabric using different tea aqueous extracts

T a b l e 2

Fabric color strength (K/S)

Fabrics	Tea aqueous extracts			
	GT	BT	RT	HT
WO	16.86	17.28	19.12	17.78
PA	7.71	8.18	8.28	11.38
CO	4.52	8.42	2.47	1.65
CA	12.23	12.96	9.39	10.11

Surprisingly, the *K/S* values listed in Table 2 are higher than those in the literature. For example, when Rehman *et al*.[37] dyed CO with BT, the calculated *K/S* values were below 1, which was overcome by fabric cationization before dyeing, resulting in deep shades (*K/S* = 8.996). The mentioned result is close to that obtained in the current study (8.42). Gorjanc *et al*.[38] improved the low affinity of GT for dyeing CO by mordanting (*K/S* of 0.61 *vs.* 10.19) with $AgNO_3$. In the recent literature,[39] even mordanting with $FeSO_4$, $CuSO_4$, or $AlK(SO_4)_2$ did not significantly improve the low *K/S* value (2.20) of wool yarns dyed with HT. All discussions confirmed that the dyeing procedure can be shortened by carefully chosen experimental conditions while avoiding using additional chemicals.

3.3. *Antioxidant and antimicrobial activity of functionalized multifiber fabrics*

Taking into account that the examined tea aqueous extracts are rich sources of polyphenols (*TPC* ranging between 2283 and 7251 mg l^{-1}), it is reasonable to assume that they can impart antioxidant activity to multifiber fabric components. Antioxidant properties are very important since they lead to reduced production of free radicals that increase oxidative stress, leading to DNA damage. Moreover, they may also contribute to the anti-inflammatory effect. As in the case of tea aqueous extracts, the fabrics' antioxidant activities were evaluated by ABTS and DPPH assay, and the comparative results are summarized in Table 3.

Except for PAN functionalized with HT aqueous extract, all other studied fabrics showed very high (81.60–100 %) ABTS radical scavenging ability, irrespective of the extract used. Five of six fabrics functionalized with GT aqueous extract possessed 100% antioxidant activity. Functionalization of PA, CO, and CA with GT aqueous extract also showed excellent ability for trapping the free radicals of oxygen species, *i.e.*, inhibition of almost all DPPH radicals present in testing solutions, Table 3.

T a b l e 3

The antioxidant activity of the functionalized multifiber fabrics determined using DPPH and ABTS methods

Tea aqueous extract	Fabrics					
	WO	PAN	PES	PA	CO	CA
	Antioxidant activity determined according to the ABTS method, %					
GT	88.47	100	100	100	100	100
BT	89.79	89.93	91.04	89.51	88.82	89.51
RT	87.22	81.60	88.06	91.18	90.63	89.86
HT	88.26	65.90	91.18	99.93	99.65	100
	Antioxidant activity determined according to the DPPH method, %					
GT	96.15	66.68	54.93	100	100	100
BT	97.27	91.58	73.98	98.36	96.32	97.14
RT	41.61	48.55	51.51	92.04	65.56	97.63
HT	23.06	23.42	20.07	100	27.20	84.57

It has to be underlined that *TPC*s have good linear correlations (r = 0.962 and 0.941) with the antioxidant activity of WO and PAN, determined according to the DPPH method. This behavior indicates that polyphenols are the most probable reason for the high antioxidant activity of the mentioned fabrics. Research conducted in the literature[40,41] showed that cotton and wool fabrics functionalized with GT aqueous extract having 90% antioxidant activities can be used for free radical elimination. However, no data is available regarding the antioxidant activities of other textile materials functionalized with GT, BT, RT, or HT aqueous extracts.

Having the tea aqueous extracts' MIC values (Table 1) in mind, the antimicrobial activities of multifiber fabric components functionalized with GT aqueous extract were tested against *S. aureus*, *E. faecalis*, *E. coli*, and *C. albicans*, while the antibacterial activities of fabrics functionalized with BT aqueous extract were tested against *S. aureus* and *E. faecalis*. The results given in Table 4 point out that all multifiber fabric components (WO, PAN, PES, PA, CO, and CA) functionalized with GT aqueous extract inhibited the growth of bacteria *S. aureus* and *E. coli* (inhibition zones between 16.0 – 21.7 mm), while PAN and CO also inhibited the growth of *E. faecalis* and *C. albicans*, respectively. In the case of WO, PAN, PES, and PA fabrics, *S. aureus* is more sensitive than the *E. coli* strain. Inhibition zones for *S. aureus* (15.3–22.3 mm) are also observed for all fabrics functionalized with BT aqueous extract, while they do not exist for the bacterium *E. faecalis*. Interestingly, the antimicrobial activity of textile materials functionalized with BT aqueous extract has not been studied yet. Studies covering the antimicrobial activity of textiles functionalized with GT aqueous extract are rare. The experiments carried out in paper[42] showed that WO fabric dyed with GT aqueous extract exerted antibacterial activities of 90% and 80% against *S. aureus* and *E. coli*, respectively. Furthermore, viscose fabrics capable of inhibiting the growth of *E. coli* and *S. aureus* were obtained by the functionalization with GT extract prepared using ethanol as a solvent.[43]

Treatment of WO, PAN, PES, PA, CO, and CA with GT, BT, RT, or HT aqueous extracts seems to be very promising for simultaneous fabric dyeing and functionalization or for only imparting antioxidant activity, *i.e.*, the ability to trap the free radical of oxygen species, preventing cell deterioration and growing a new cell in the skin. Due to the different manifestations of antioxidant and antimicrobial activity, fabrics of different chemical compositions functionalized with GT or BT aqueous extract can be considered high-value-added medical textiles having therapeutic, prophylactic, and protective functions.

Table 4

The width of the inhibition zone (clear zone), mm; / – no effect

Fabrics	Tea aqueous extract					
	GT				BT	
	S. aureus	*E. faecalis*	*E. coli*	*C. albicans*	*S. aureus*	*E. faecalis*
WO	20.0	/	17.7	/	19.3	/
PAN	18.0	16.0	16.7	/	17.0	/
PES	21.7	/	21.0	/	17.5	/
PA	21.7	/	18.3	/	15.3	/
CO	17.7	/	17.7	18.7	22.3	/
CA	16.0	/	18.7	/	15.8	/

4. CONCLUSION

The GT, BT, RT, and HT aqueous extracts can be efficiently exploited for dyeing wool, cellulose acetate, polyamide, and cotton. This is confirmed by the fabrics' color strength values of 1.65–19.12. The studied tea aqueous extracts had 415–1213 mg l^{-1} and 2283–7251 mg l^{-1} *TFC* and *TPC*, respectively. Depending on the test method used, DPPH or ABTS, their antioxidant activities are in the range of 78.1–93.1 % or 97.8–100 %, respectively. The MIC value of 500 μg ml^{-1} was found against *S. aureus* and *E. faecalis* for both BT and GT aqueous extracts, while GT aqueous extract additionally exhibited the same activity against *E. coli* and *C. albicans*. Due to the above-mentioned extracts' properties, WO, PAN, PES, PA, CO, and CA functionalized with GT aqueous extract inhibited the growth of bacteria *S. aureus* and *E. coli*, while PAN and CO also inhibited the growth of *E. faecalis* and *C. albicans*, respectively. Wide inhibition zones for *S. aureus* were observed for fabrics functionalized with BT aqueous extract. Generally, the studied fabrics showed very high (81.60–100 %) ABTS radical scavenging ability independent of the extract used. It can be concluded that fabrics with different chemical compositions dyed and/or functionalized with GT or BT aqueous extract can be employed to produce high-value-added medical textiles with therapeutic, prophylactic, and protective functions. They can find potential applications in wound treatment, especially in skin wounds that are susceptible to infection with *S. aureus*. Moreover, WO and CO functionalized with GT or BT aqueous extracts can also be considered for disposable medical textiles like bandages and gauze used in the wound-healing process.

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