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One-Pot Robust Dyeing of Cotton Fabrics with Multifunctional Chamomile Flower Dyes

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Abstract: Our study focused on the extraction of biologically active compounds in the form of eco-friendly dyes from dried chamomile flower (CF) powder using three different solvents, as well as the development of an approach to the simultaneous dyeing and finishing of cationized cotton fabrics, a process which can impart multifunctional properties. These extracted dyes were successfully used as reducing and stabilizing agents in the synthesis of silver nanoparticles (AgNPs), which functionalized both the dye and the fabric. This green, value-added dyeing approach is needed for the production of biomedical textiles, especially in the healthcare sector. Our results revealed that the total phenolic content (TPC) and total flavonoid content (TFC) were significantly higher in dyes extracted using ethanol and methanol than those extracted using an aqueous solution. Furthermore, cotton fabrics dyed with these three dyes exhibited a wide range of colors with good washing fastness, excellent UV protection, and antioxidant properties. The simultaneous dyeing and finishing of cotton fabrics via the in situ green synthesis CF extracts/AgNPs dyes enhanced the *K/S* values and the antimicrobial, antifungal, and antioxidant activity of the fabrics while still providing adequate UV protection. This process also improved the washing fastness and durability, which is important for biomedical applications.

Keywords: Chamomile flower, Dye extraction, Silver nanoparticles, Cotton dyeing, Multifunctional properties

Introduction

In modern society, many peoples choose to lead healthy lifestyles by using natural materials such as herbs (plants) for applications such as food colorants, cosmetics, coloration, and other functions employed in textile materials. Nevertheless, the coloration of textiles with synthetic dyes and their toxic auxiliaries are of major global interest, but the use of synthetic dyes can harm both humans and the environment [1]. Hence, due to the increasing awareness of ecological and health problems associated with synthetic dyes, comprehensive studies must be conducted to revitalize the use of natural dyes in the textile industry. Another key reason to use natural dyes is their multifunctional nature; they can possess antimicrobial, antioxidant, insect repellent, flame retardant properties, among others, as well as provide protection from UV light. In addition, they can imbue the dyed fabric with unique and elegant colors of different hues and tones [2,3].

Chamomile is the most popular aromatic medicinal plant globally and is extremely popular due to its therapeutic capabilities. Its dried flowers and essential oils have a wide range of pharmaceutical and cosmetic applications because it exhibits several medicinal properties, such as antimicrobial, anti-inflammatory, antibiotic, antioxidant, analgesic, and antiallergic capabilities, among others [4,5]. The sustainable production and efficiency of chamomile flower (CF) have made their extracts the subject of global attention. The CF extract is rich in bioactive compounds such as phenolics and flavonoids, such as apigenin, quercetin, patuletin and luteolin glucosides, and coumarins. In addition, its essential oils provide antioxidant, antimicrobial, anti-inflammatory, moisturizing and medicinal properties [6-8]. At present, the most efficient isolating techniques of the phytochemicals contained within chamomile extracts in particular, the polyphenolic compounds generally involve organic solvents such as ethanol, methanol, and glycerol as well as aqueous solutions using conventional, ultrasound and microwave-assisted extractions, Soxhlet, and supercritical water extraction [9]. Previous textile researchers have utilized the aqueous extract of CFs as natural dyes in the coloration of wool fabrics to investigate the kinetic models that govern the dyeing mechanism; these were found to be consistent with a pseudo-second-order reaction and the Langmuir-Freundlich isotherm [10]. In addition, dyeing fabrics with red onion, madder, chamomile and red onion peel/chamomile aqueous extracts with or without mordants has been reported to improve the fastness of the fabric and confer improved UV protection capabilities [11].

Recently, the modern medical textile industry has investigated nanoparticulate technology in the application of colorants. These colorants do not contain any chromophores in their chemical structure; their color is solely due to surface plasmon resonance, size, and shape [12]. Currently, the most commonly used coloring process is the chemically toxic reduction-oxidation reaction between a salt precursor and a reducing agent. These chemical reagents are expensive and have potentially severe environmental impacts. Hence,

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the use of plant extracts is generally preferred because the synthesis of plant-mediated nanoparticles is simple, cost-effective, eco-friendly, and less hazardous for humans. However, the reaction mechanisms of these green synthesis methods must be studied due to the presence of an enormous number of bioactive compounds in the plant extract as well as the wide variety of plants species. The bioactive compounds within the aqueous extracts of CFs have been widely utilized in the biosynthesis of various nanoparticles such as magnesium oxide, manganese dioxide, zinc oxide, and silver nanoparticles. These compounds were synthesized via green approaches and have been evaluated as promising antibacterial agents [13-15]. In particular, silver nanoparticles (AgNPs) are widely used due to the broad spectrum of their antimicrobial potential; they have been integrated into numerous products such as wound dressing agents as well as antimicrobial coating agents in medical devices that are used to sterilize air, textiles and surfaces [16]. Furthermore, the biosynthetic method in which AgNPs are produced is more desirable than other physicochemical techniques as they can be mass produced while being safely utilized for various therapeutic applications. In addition, AgNPs are very frequently integrated into the textile dyeing process using plant extracts [2,17,18].

In this context, the development of biomedical dyed textiles through the production of cellulose-based technical textiles such as wound dressings, kenezue, bandages, and antibacterial masks has gained significant momentum in the healthcare sector. In particular, the manufacture of bioactive functionalization of cotton textile fabrics has developed into a matter of strategic importance. Many researchers have used antimicrobial natural dyes as an efficient means of integrating the dyeing and finishing process into a single bath to increase its potential applications as well as to minimize its environmental impact through an eco-friendly cost-effective methodology [1,16,19]. At present, many researchers have been motivated to develop numerous strategies for the in situ generation of AgNPs on cotton fabrics due to their effective antimicrobial capabilities [2,12,18].

The fundamental aim of this study was the green synthesis of multifunctional dyes from CF extracts. It is also aimed to synthesize and dye cotton fabrics with CF/AgNPs dyes via a one-pot dyeing process to produce multifunctional cotton fabrics that are suitable for use in the healthcare sector. To accomplish this, natural colorants were extracted from CFs through the use of three solvents: methanol, ethanol, and an aqueous solution. To obtain the highest yield of specialized bioactive compounds, the optimal extraction conditions for each solvent was investigated. The structure of the extracted dyes was characterized and analyzed, and their total phenolic content (TPC) and total flavonoid content (TFC) were determined. The extracted dyes were used as an effective reducing and stabilizing or chelating agent for the biosynthesis of AgNPs. Hence, the in situ dyeing of cotton fabrics with the functional CF/AgNPs dyes via a one-pot dyeing process was used to impart the fabric with multifunctional properties. The dyed fabrics were subjected to, X-ray diffractometer (X-ray), scanning electron microscopy (SEM) and Energy dispersive X-ray (EDX), antimicrobial, antioxidant, and ultraviolet protection factor (UPF) analysis to identify the functional groups contained within the dye components as well as study the morphology and fastness properties of the dyed fabrics. The colorimetric data, color strength (*K/S* values) and washing fastness of the dyed fabrics were also studied.

Experimental

Materials

Dried chamomile flower powder was purchased from a local market in Egypt. Bleached cotton fabric was kindly supplied by the Misr Company for Spinning and Weaving, Mahala el-Kubra, Egypt. Methanol, ethanol, citric acid, sodium hydroxide, sodium hypophosphite (SHP), and silver nitrate were obtained from Fluka. Polyethyleneimine (PEI) with a molecular weight of 60,000 was acquired from Sigma-Aldarich, Germany. All chemicals were used in this study without further purification. Cotton fabric was washed in a 5 g/*l* non-ionic detergent solution (Hostapal CV, Clarient) at a 1:50 material: liquid ratio at 95 °C for 4 h to remove any impurities. The fabric was then thoroughly rinsed with tap water and dried at room temperature.

Extraction of Dyes from Chamomile Flowers

Dyes were initially extracted from CF powder by simple shaking it in a water bath using methanol, ethanol and an aqueous solution as solvents. 5 g of CF powder dissolved in 100 m*l* from each solvent in quick-fit stainless steel reaction vessels. To optimize the extraction process, different parameters such as solvent strength (40-100 %), extraction temperature (40-100 °C), extraction time (15, 30, 45, 60, 90, and 120 min), and the amount of CF powder used were varied during the extraction to investigate the combination of parameters that would maximize the dye extract yield and/or solid content percentage (SC%). The liquid extract was filtered through Whatman filter paper No.1 using vacuum filtration. The filtrate was then centrifuged at 4000 rpm for 15 min to remove any trace of the residue. All the liquid extracts were evaporated at room temperature and the extracted dyes (in powder form) were stored at 4 °C for further analysis. The SC% was determined gravimetrically by taking a known volume of filtrate extracts, evaporating them under vacuum, and drying them in a vacuum oven at 105 °C until a constant weight was obtained. Each analysis was performed in triplicate and the SC% was calculated in

terms of percentage weight (w/w) [20].

Cationization of the Cotton Fabric

The cotton fabrics were cationized with PEI in both the absence and presence of a crosslinking agent [21]. Cotton fabrics were activated by immersing them in 1 N sodium hydroxide solution at a 1:50 material: liquid ratio. They were then shaken for 15 min at room temperature, squeezed, rinsed with distilled water, and dried at room temperature. In samples without a crosslinking agent, the activated cotton fabrics were then immersed in an aqueous solution of 10 % PEI at room temperature for 30 min, squeezed, and washed with distilled water to remove any remaining unbonded PEI solution, before being dried at room temperature. In samples with a crosslinking agent, the activated cotton fabrics were immersed in a mixture containing an aqueous solution of 10 % PEI mixed with 10 g/*l* of citric acid and 5 g/*l* of SHP. The fabric was subsequently padded to pick-up 80 % twice, and dried in an oven at 70 °C for 3 min. It was then cured at 140 °C for 3 min, washed with distilled water, and dried at room temperature [22].

Dyeing Cotton Fabrics with Dyes Extracted from CF in Different Solvent

The cationized cotton fabrics were dyed in the presence or absence of crosslinking agent with ethanol extracted CF (CFEE), methanol extracted CF (CFME) and aqueous extracted CF (CFAE) dyes. This was done by using a 1:50 ratio of material to dye solution at 95 °C for 60 min. Each dye solution had a constant concentration of 10 % dye powder (w/v), extracted using their respective solvent. At the end of the dyeing process, the dyed fabrics were rinsed with hot water several times, followed by rinsing in tap water, followed by a final 30 min wash in a solution containing 3 g/*l* non-ionic detergent (Hostapal CV, Clarient) at 60 °C. Finally, the dyed fabrics were thoroughly rinsed with water and dried at room temperature. For the in situ dyeing of the fabrics using CF/AgNPs (CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs), the dye solution was used as a reducing and stabilizing agent for 0.1 mM silver nitrate solution. Different volumes of $AgNO_3$ solution were added dropwise to the dye bath solution while being thoroughly stirred at 75 °C for 15 min. 1 g of cationized cotton fabric was then dyed in this dyebath solution at 75 °C for 30 min, following which the temperature was gradually raised to 95 °C and held for 60 min. At the end of the dyeing process, the dyed fabrics were rinsed in hot water and tap water, before being squeezed and washed for 30 min in a bath containing 3 g/*l* non-ionic detergent at 60 °C. The dyed fabrics were then rinsed with tap water and air-dried. To find the optimal parameters for the biosynthesis of AgNPs dyes, the $AgNO_3$ concentration (170-5440 mg/*l*), temperature range (75-95 °C) and reducing time (5, 10 and 15 min) were varied for each experiment.

Extraction Characterization Methods

Analysis of UV-Visible Spectra

The UV-Visible spectra (200-800 nm) of the dye solutions extracted from CF using different solvents were examined and analyzed using a UV-Visible absorption spectrophotometer (JASCO, Japan).

Analysis of FTIR Spectra

The FTIR spectra of the extracted dyes were analyzed on an FTIR spectrophotometer (JASCO, Japan) between 4000-500 cm^{-1} to identify the functional groups present in the bioactive components of each CF dye.

Analysis of Total Phenol Content and Total Flavonoid Content

The TPC and TFC of each CF dye were determined spectrophotometrically by the Folin-Ciocalteu colorimetric method and the aluminum chloride ($AlCl_2$) method, respectively [23,24].

Dyed Fabric Characterization Methods

Analysis of the UV-Visible Spectra of the Dyed Fabrics

The UV-Vis absorption spectra of the dyed cationized cotton fabrics were analyzed using an Ultra Scan PRO spectrophotometer (Hunter Lab, USA) between 350-700 nm.

SEM and EDX Analysis

The surface morphology of the dyed cotton fabrics was investigated by scanning electron microscopy (SEM) using a HITASHI S-3000 microscope with an acceleration voltage of 15 kV. Each sample was coated with thin film of gold before analysis. Their chemical analysis was measured by surface energy dispersive X-ray spectroscopy (EDX) attached with the same microscope.

XRD Analysis

X-ray diffraction was conducted using an Empyrean (Panalytical Co., New Zealand). The instrument utilized nickel-filtered Cu-Kα radiation (1.54060 Å), with incident radiation of 45 kV and 30 mA. The diffracted intensities were recorded between angles of 4-80 ° 2θ at a scanning speed of 3 °/min.

Colorimetric Measurement

The colorimetric parameters of the dyed fabrics including lightness (L^*), red/green coordinate (a^*), and yellow/blue coordinate (b^*) as well as its *K/S* values were evaluated using UltraScan PRO spectrophotometer (Hunter Lab, USA).

Color Fastness in Terms of Washing Durability

The color fastness of the cotton fabrics with respect to washing durability for each washing cycle up to 5 cycles were evaluated according to the ISO 105-C10:2006 standard.

Measurement of Ultraviolet Protection Factor

The UV protection factor (UPF) of the undyed and dyed fabrics were measured according to the AATCC Test method 183:2010 by using a U-750 spectrophotometer (JASCO, Japan).

Evaluation of Antibacterial and Antifungal Activity

The antimicrobial activity of the undyed and dyed cotton

fabrics was evaluated against gram-positive bacteria (*Bacillus subtilis* and *Staphyllococus aureus*), gram-negative bacteria (*Escherichia coli* and *Pseudomonas aeruginosa*), and fungi (*Candida albicans*) using a quantitative method; the results are presented in reduction percent [25].

Evaluation of Antioxidant Activity

The antioxidant activity of the dyed cotton fabrics was investigated by assessing their free radical scavenging ability using a 2,2-diphenyl-1-picrylhydrazyl DPPH assay [26].

Results and Discussion

Effect of Solvent and the Optimization on the Extraction Process

Considering that the extraction of natural dyes from plant material is an extremely important stage in the isolation and identification of bioactive compounds, optimizing the extraction parameters has been the focus of many studies that seek to perfect the dye extraction process [27,29]. The extraction efficiency of ethanol, methanol, and aqueous solutions was investigated by varying different parameters such as solvent strength, extraction temperature, extraction time, and CF concentration to determine the maximum absorbance (A) and solid content (SC%) for each solvent.

Figure 1(A) reveals that increasing the strength of the ethanol and methanol hydroalcoholic solutions from 40-80 % increased dye absorbance and SC%. Increasing the strength of both these solvents could result in a greater degree of dye transfer from the CF powder (solid mass) to the liquid surface (solvents), maximizing the SC% [28,29].

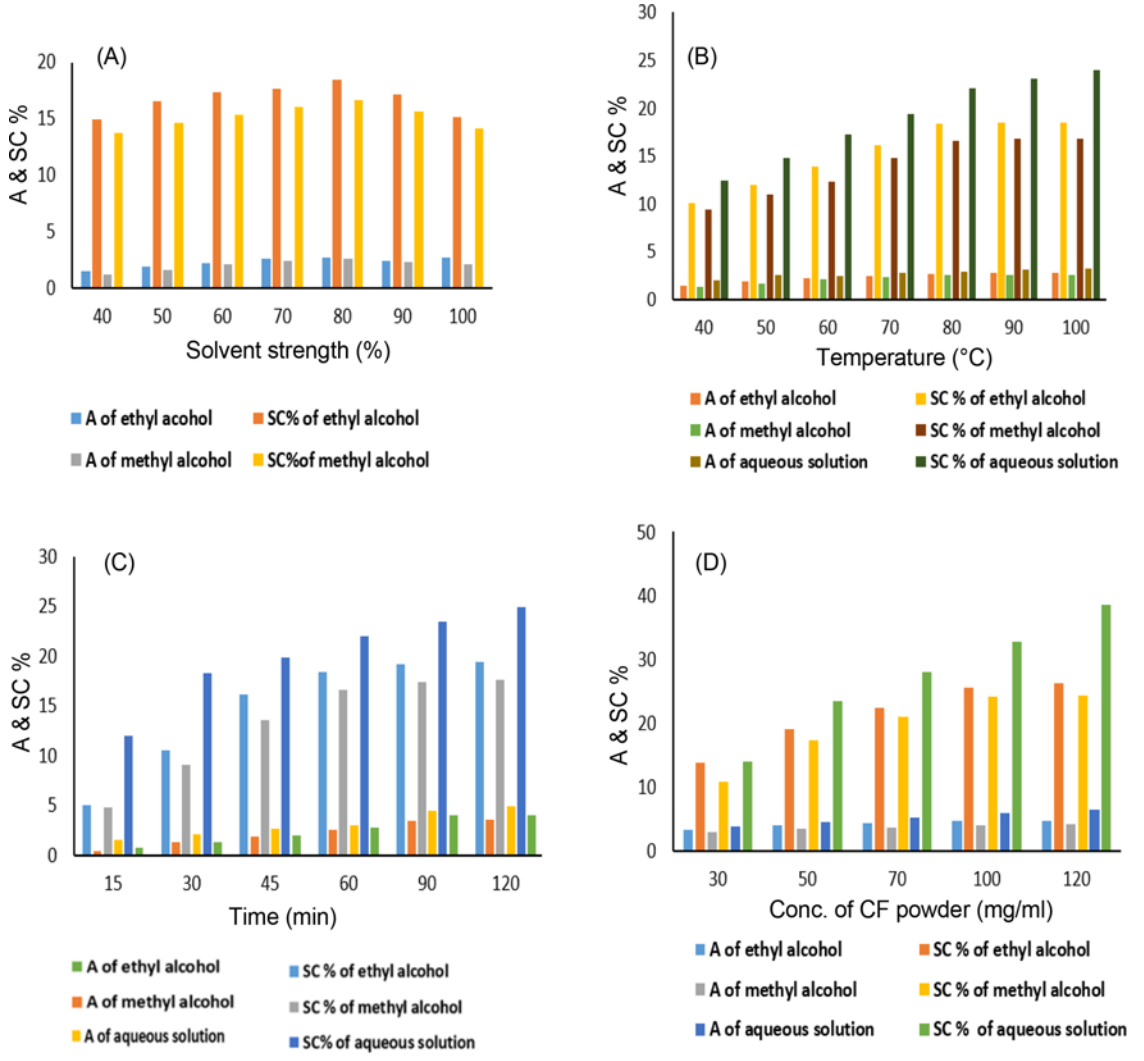


Figure 1. Effect of solvent and optimization of CF dye extraction in relation to the absorbance (A) and solid content percent (SC%). (A) effect of solvent strength (50 mg/m*l* concentration of CF powder at 80 °C for 60 min), (B) effect of temperature (50 mg/m*l* of CF powder, 80 % solvent strength for 60 min), (C) effect of time (50 mg/m*l* of CF powder, 80 % solvent strength at 80 °C) and (D) effect of different concentration of dye (80% solvent strength at 80 °C for 90 min).

However, above a hydroalcoholic strength of 80 %, the A and SC% of the extracts decreased due to the decreasing polarity of the solvent.

Figure 1B shows that the A and SC% of CF dyes extracted with 80 % ethanol, 80 % methanol, and the aqueous solution extract increased significantly when the extraction temperature was increased from 40 to 80 °C. Increasing temperatures lead to an increase in the diffusion coefficient, which consequently results in the rapid diffusion of dye components from the solid CF mass into the liquid solution. However, above 80 °C, there is only a slight increase in A and SC% before it plateaus; this could be attributed to internal redox reactions, hydrolysis and polymerization [30-32].

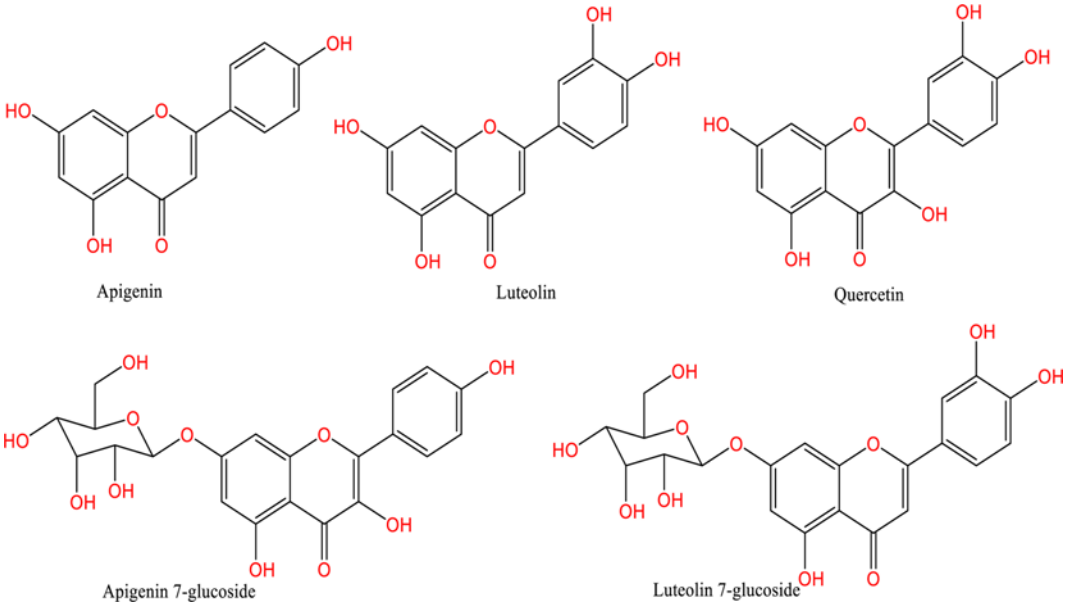
In contrast, Figure 1(C) clearly shows that the A and SC% of each extracted dye increased as the extraction time was increased from 15 min to 90 min. This was because longer extraction times allow the solvents to swell and moisturize the plant, solubilizing the phenolic compounds inside vegetable cells, and allowing them to diffuse into the solvent, thereby increasing the extraction rate [30-32]. Beyond 90 min, only a slight increase was observed in both the A and SC% of the dyes extracted with all three solvents, indicating that an approximate equilibrium had been reached. This could be due to the saturation of each solvent (80 % ethanol, 80 % methanol, and the aqueous solution) during the extraction of active phenolic dye compounds.

Figure 1(D) also shows that as the concentration of the CF powder is increased from 30-120 mg/m*l* during the extraction process, the A and SC% of the dyes extracted by all three solvents significantly increased. This suggests that increasing the concentration of CF powder also increased the availability of soluble solids and polyphenolic compounds during the extraction process, which prevent the solvent from becoming saturated. Furthermore, above a CF concentration of 100 mg/m*l*, the dyes extracted with 80 % ethanol and methanol appear to become saturated with phenolic compounds, slightly increasing the SC%. However, both the A and SC% of the dyes extracted using the aqueous solution increased linearly as the concentration of CF powder was increased from 30-120 mg/m*l*. This suggests that the aqueous solution was not saturated, allowing for the extraction of more soluble solids (such as some phenolics and other compounds).

Characterization of CF Extracted Dyes in Different Solvents

UV-Visible Characterization

The phenolic compounds present in the CF extracted dyes under optimal extraction conditions (80 % ethanol, 80 % methanol, and aqueous solutions extracted at 80 °C for 90 min using 100 mg/m*l* amount of CF powder) were determined by an analysis of the UV-Vis absorbance spectrum as shown in Figure 2. The dye extracted using the aqueous solution exhibited two broad peaks; the higher peak at 265 nm indicates the presence of auxochrome and chromophore substituents in the aromatic phenolic structure while the lower, broader peak at 310-360 nm represents a mixture of some water-soluble flavonoids compounds [2,10]. In contrast, the CFEE and CFME dyes exhibited three sharp peaks; the first peak appeared at 275-285 nm and was attributed to the n-π* transitions of the substituents (OH, COOH, and C=O) present in the aromatic skeleton of the phenolic components [2]. In addition, both dyes exhibited two distinct peaks at 320-330 nm and 375 nm that indicate a



Scheme 1. Chemical structure of the main flavonoids chromophores present in CF dyes.

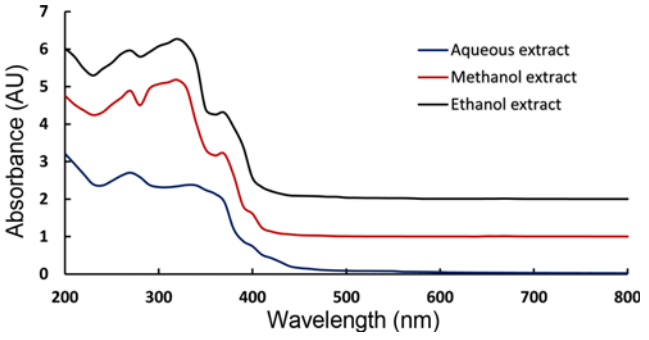


Figure 2. UV-visible spectra of the CF dye extracts in three different solvents.

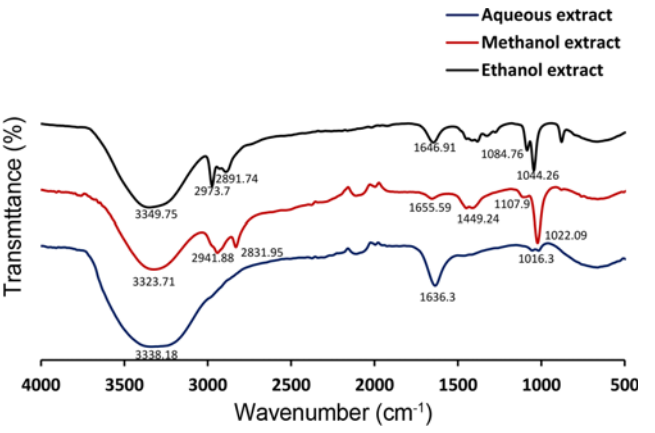


Figure 3. FTIR spectra of CF extracted dyes in different solvents.

high concentration of flavonoids [33-37]. Among the main flavonoids present in CF, apigenin and its derivatives may be the dominant chromophore compounds associated with all the three extracted dyes. The structure of some main chemical components present in CF dyes is presented in Scheme 1.

Figure 2 suggests that the CFEE and CFME dyes were more efficient at extracting the phenolic and flavonoid compounds due to their higher polarity and solubility than the CFAE dye [38,39].

FTIR Analysis

The FTIR spectra of the three dyes are shown in Figure 3. The spectrum of the dye extracted using the aqueous solution exhibited a large, intense peak at 3338.18 cm^{-1}; this was attributed to the -OH stretching vibrations of phenolic rings. The peak at 1656.35 cm^{-1} was attributed to the stretching vibration of -C=O, and the peak at 1016.3 cm^{-1} may be associated with the deformation vibration of -OH [40]. In contrast, several bands were observed in the spectrum obtained from the dye extracted using methanol, the strongest absorption peak at 3323.71 cm^{-1} was attributed to the -OH stretching vibration of the phenolic compounds associated with the intermolecular hydrogen bonded framework. Peaks at 2941.88 cm^{-1} and 2831.95 cm^{-1} were attributed to the stretching vibration of CH_2 and CH_3 groups attached to the aromatic ring, respectively. The band at 1655.59 cm^{-1} may be due to the stretching vibration of C=O in conjugated aromatic compounds (such as ketones, aldehydes, quinones, and esters) present in the phenolic and flavonoid compounds. Another band at 1449.24 cm^{-1} refers to the C-C stretching vibrations of aromatic rings, while peaks at 1107.9 and 1022.09 cm^{-1} correspond to C-O stretching vibrations. Figure 3 shows that the dye extracted with ethanol exhibited several bands that were similar to those observed in the methanol extracted spectrum due to the presence of OH, CH, C=O, C=C, and C-OH groups from the flavonoids or phenolic compounds present in the plant material. Peaks appearing at 1084.76 cm^{-1} and 1044.26 cm^{-1} may be due to alkoxy groups [41,42].

Total Polyphenol and Flavonoid Content Analysis

Polyphenolic compounds and flavonoids are secondary metabolites of plants; these compounds exhibit a wide range of chemical structures and bioactivities. Table 1 shows that the dye with the highest extraction yield in percent was CFAE. This suggests that yield percent was not only related to the TPC and TFC but is also associated with other water soluble organic compounds present in CF. In contrast, CFEE and CFME exhibited the highest TPC values (1523.80 µg GAE/m*l*, and 1398.62 µg GAE/m*l*, respectively) and TFC values (845.91 µg CE/m*l* and 678.75 µg CE/m*l*, respectively) compared to the lower values exhibited by CFAE (1018.00 µg GAE/m*l* and 444.11 µg CE/m*l*). The results of this phytochemical study may be due to the hydrolysis of the conjugated phenolic compounds, which were easily extracted using 80 % ethanol and 80 % methanol due to their high polarity and a greater degree of solubility [38,39].

Optimization of the Dyeing Process

The results of this study indicate that the chemical composition of phenolic and flavonoid compounds present in the three extracted dyes may adversely affect the functionality of these dyes as a coloring agent in the dyeing of cotton fabrics. However, these CF dyes could play an essential role in the green reduction of silver ions (Ag^{+}) to stable AgNPs (Ag°). Furthermore, the synthesis of CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs dyes could be

Table 1. Extract yield, total phenolics (TPC), and total flavonoids (TFC) content of CF dye extracted in different solvent

Dye extract	Dye extract yield (%)	TPC (µg GAE/m*l*)	TFC (µg CE/m*l*)
Aqueous extract	32.80	1018.00	444.11
Methanol extract	24.29	1398.62	678.75
Ethanol extract	25.60	1523.80	845.91

used as a metallic mordant to increase the functionality and fixation of the extracted dyes while also imparting the cationized cotton fabric with multifunctional properties that have a wide variety of applications in the biomedical textile sector. Consequently, optimal extraction conditions are required to synthesize AgNPs dyes and optimize the dyeing conditions of the CF extracted dyes on cotton fabrics. To optimize the synthesis of AgNPs and the dyeing process, several parameters were studied to obtain the maximum *K/S* values in presence and absence of AgNPs dyes in an eco-friendly approach to augmenting a dyed fabric with multifunctional properties for use as a biomedical textile.

Table 2 demonstrates that, when dyeing the cationized cotton fabric with CFEE/AgNPs, the *K/S* values increased significantly as the concentration of $AgNO_3$ was increased to 5440 mg/*l*. This result was attributed to the increase in the concentration of the bioactive components in the CFEE as well as the concentration of $AgNO_3$, which consequently increased the in situ growth of AgNPs on the fabric and caused the color to shift to a darker shade.

In contrast, a maximum *K/S* values was obtained when the concentration of $AgNO_3$ was increased to 340 mg/*l* when dyeing cotton fabric with both CFME/AgNPs and CFAE/AgNPs. This may be because the bioactive phenolic extracted dye components of CFME and CFAE were completely exhausted in the reduction process of Ag^+ in $AgNO_3$ to Ag° NPs, resulting in the equilibrium deposition of AgNPs. Above 340 mg/*l*, the large amount of AgNPs deposited on the fabric increases the chance of collisions between AgNPs, increasing their kinetic energy; this results in aggregation followed by the precipitation of these nanoparticles in the dye bath solution and produces colors with a lighter shade.

However, dyeing the cationized cotton fabric with CFEE/AgNPs, CFME/AgNPs, and CFAE/AgNPs in the presence of crosslinking agent exhibited maximum *K/S* values only when the concentration of $AgNO_3$ was increased to 5440 mg/*l*. As the concentration of AgNPs in the dyed fabric increased, they were chelated and stabilized due to the presence of both OH phenolic groups of CF extracted dye as well as the COOH groups in the crosslinking agent of cationized cotton fabric [43,44]. Table 3 shows that increasing the temperature of the reduction process from 75-95 °C was not associated with any significant increase in the *K/S* values of the dyed fabrics. This may be because the deposition of the three AgNPs dyes onto the fabric reaches an equilibrium at 75 °C.

The effects of increasing the duration of the reducing process from 5-15 min on the *K/S* values of the dyed cationized cotton fabric in the presence and absence of the

Table 2. Optimization of synthesis of AgNPs and dyeing process as a function of $AgNO_3$ concentration

Conc. of $AgNO_3$ (mg/*l*)	*K/S* of cationized cotton fabric dyed with:			*K/S* of cationized cotton fabric+crosslinking agent dyed with:		
	CFEE/AgNPs	CFME /AgNPs	CFAE/AgNPs	CFEE/AgNPs	CFME/AgNPs	CFAE/AgNPs
0	10.01	15.98	11.13	13.69	16.41	14.82
170	22.14	14.34	14.00	-	-	-
340	10.39	23.54	16.23	14.49	17.68	16.35
680	10.82	21.31	16.03	-	-	-
1360	12.21	11.63	15.78	16.45	18.94	16.50
2040	14.62	7.95	15.60	-	-	-
2720	15.51	7.80	-	23.01	19.99	17.02
3400	17.39	7.50	15.04	23.79	22.11	17.36
4080	18.72	-	-	27.73	24.00	17.71
5440	24.63	-	-	34.80	26.58	18.15

Table 3. Optimization of synthesis of AgNPs and dyeing process as a function of reducing temperature and reducing time

Sample	*K/S* of reducing temperature		*K/S* of reducing time		
	75 °C	95 °C	5 min	10 min	15 min
Cationized cotton fabric dyed with:					
CFEE/AgNPs	24.63	25.14	23.50	23.85	24.63
CFME/AgNPs	23.54	24.02	16.71	18.02	23.54
CFAE/AgNPs	16.23	16.42	14.20	15.45	16.23
Cationized cotton fabric+crosslinking agent dyed with:					
CFEE/AgNPs	34.80	35.01	21.94	29.58	34.80
CFME/AgNPs	16.58	17.23	13.56	14.70	26.58
CFAE/AgNPs	18.15	19.11	12.84	15.03	18.15

crosslinking agent is also presented in Table 3. It reveals that the *K/S* values increased markedly as the time of the process was increased. Increasing the time spent in the dye bath solution allowed the cationized cotton fabric to be impregnated with additional Ag^+ ions, increasing the in situ growth and/or the deposition of AgNPs.

Mechanism of the Synthesis and Dyeing of AgNPs onto Cotton Fabric

It was found that CFEE, CFME, and CFAE serve as anionic dyes that bear a negative charge in their solutions. However, the presence of negatively charged hydroxyl groups on the surface of the cotton fabric resulted in a repulsive force between the two materials. Hence, these phenolic dye molecules had a low affinity with the cotton fabric, and direct dyeing lead to poor fastness properties, which manifested in *K/S* values of 1.23, 0.97, and 0.34 for the CFEE, CFME, and CFAE dyes, respectively. In contrast, when the cotton fabrics were cationized with PEI to generate an active cationic side (primary and secondary amines), which act as a coating layers on the surface of the fabric as well as increase the dyeing affinity of the three dye extracts in the absence or presence of a crosslinking agent [21,45] as shown in Schemes 2(A) and 2(B). The inadequate washing fastness of the dyed fabric can be noticeably improved by complexation with metal ions.

Although the phenolic dyes extracted from CFs exhibit powerful antioxidizing properties, they have been widely used as reducing and stabilizing agents for the synthesis of nanoparticles (Scheme 2(C)). In particular, AgNPs are widely used in healthcare products such as medical textiles, wound dressings, and cosmetics; they exhibit excellent antibacterial and antifungal properties [45]. We propose that the mechanism behind the biosynthesis of CFEE/AgNPs, CFME/AgNPs, and CFAE/AgNPs dyes is as follows (Scheme 2(D)):

1) The Ag^+ ions in silver nitrate solution form an intermediate complex with the adjacent OH groups in the phenolic dyes.

2) The Ag^+ ions are then reduced to AgNPs by accepting electrons from the electron donating hydroxyl groups (-OH) in the phenolic compounds, converting the phenolic compounds to their quinone form.

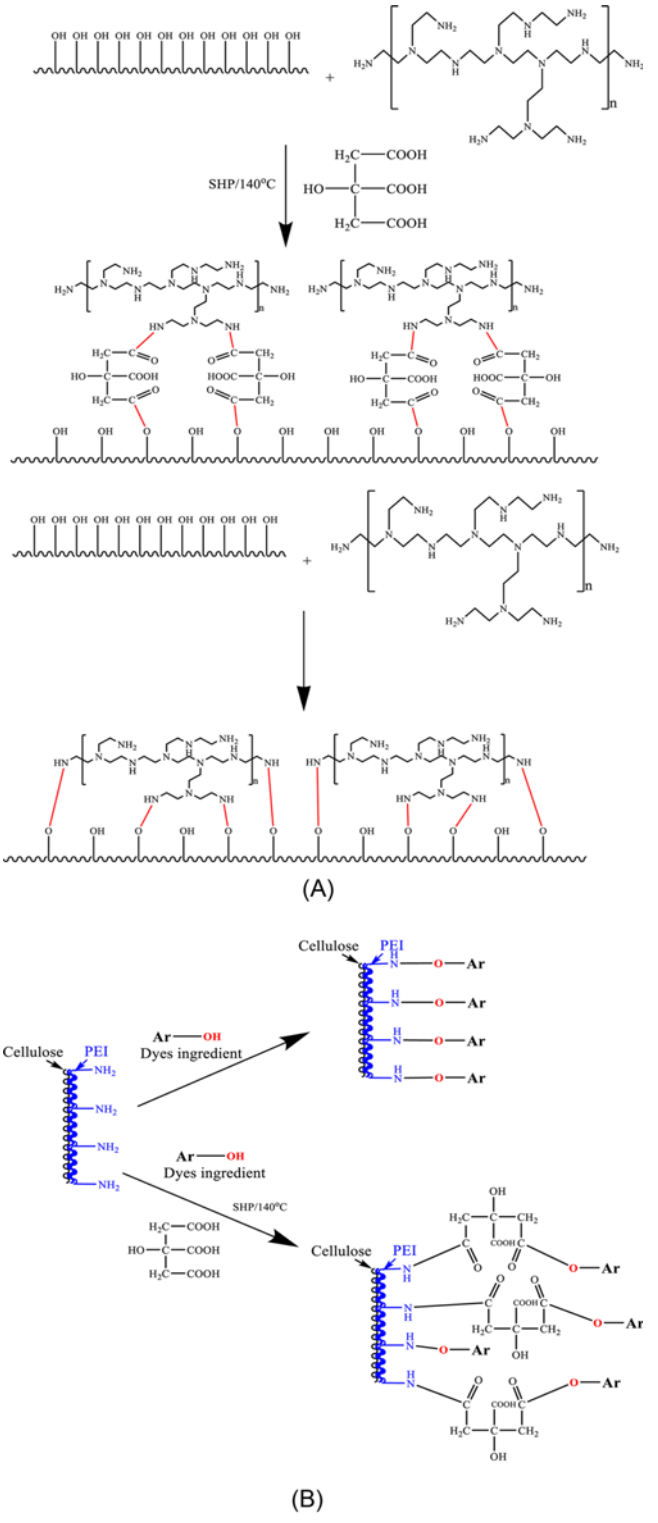
3) The quinones in the phenolic extracted dyes are adsorbed onto the surface of the AgNPs, stabilizing them.

4) During the dyeing process, the AgNPs are anchored and stabilized through the amine (primary and secondary) and/or -COO groups or both of them, whatever applying the crosslinking and cationizing agents.

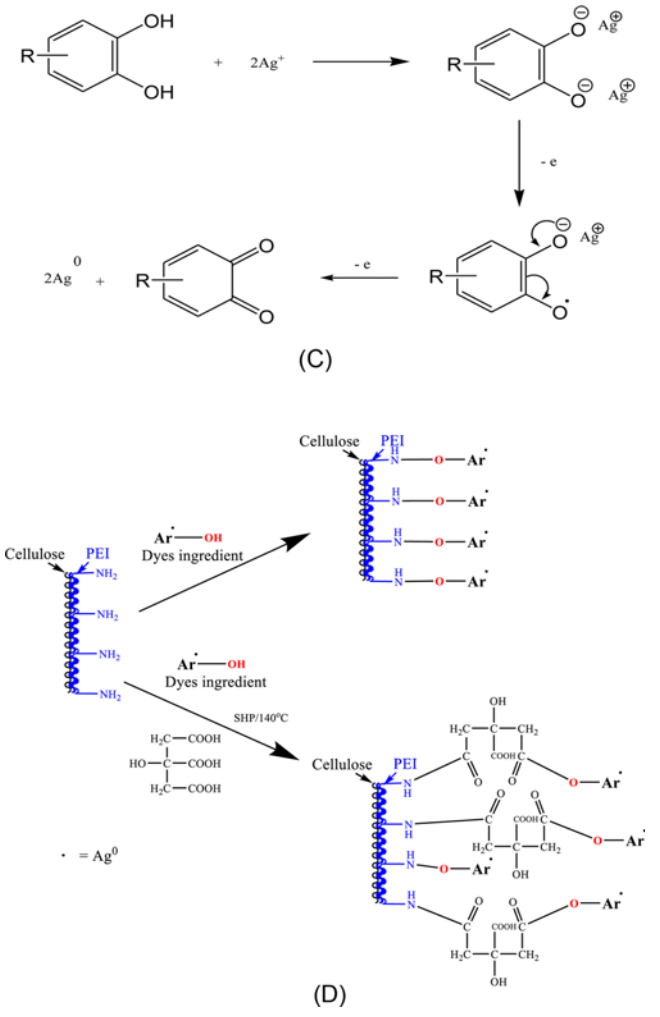
Characterization of the Dyed Cotton Fabrics

UV-Visible Characterization of the Dyed Fabric

The UV-Vis spectrum of the CF extracted dyes and the effect of CF/AgNPs dyes on *K/S* values of the dyed fabrics



Scheme 2. Mechanism of dyeing process; (A) cationization of cotton fabric in prensence and absence of crosslinking agent, (B) cationized cotton fabric dyed with CF extracts in different solvent, (C) green synthesis of AgNPs, and (D) cationized cotton fabric in the presence and absence of crosslinking agent dyed with CF/AgNPs dyes.



Scheme 2. Continued.

are evaluated in Figure 4. The spectra of cationized cotton fabric dyed with CFEE and CFME exhibit maximum *K/S* value peaks at 375 nm, while the same peak appears at 365 nm for CFAE. When the dyeing process was conducted with CFEE/AgNPs, the maximum *K/S* value peaks observed in the spectra were shifted to longer wavelengths 410 nm (red shift) as the concentration of $AgNO_3$ solution was increased from 0-5440 mg/*l*; this was attributed to the absorption band of the surface plasmon of AgNPs as shown in Figure 4(A). In contrast, cotton fabrics dyed with CFME/AgNPs and CFAE/AgNPs exhibited the higher *K/S* value peaks at 410 nm and 405 nm, respectively, when a 340 mg/*l* $AgNO_3$ solution was used (Figures 4(B) and 4(C)). This suggests that the phenolic compounds in both CF dyes were exhausted in the process of reducing and stabilizing the Ag^+ ions at a concentration of 340 mg/*l*, promoting the formation of a high concentration AgNPs on the dyed surface of the fabric. However, when the $AgNO_3$ concentration was increased beyond 340 mg/*l*, the *K/S* values of the absorbance peaks dramatically decreased. This can be attributed to the lack of the additional stabilizing and reducing agents; hence, a lower concentration of AgNPs was obtained on the dyed fabric. Alternatively, this may have increased the amount of AgNPs aggregation, which cause the AgNPs to precipitate and settle at the bottom of the dyeing bath [43,44,46]. However, dyeing the cationized cotton fabric with CFEE/AgNPs, CFME/AgNPs, and CFAE/AgNPs in the presence of a crosslinking agent confirmed the presence of AgNPs in all concentrations of $AgNO_3$ from 340-5440 mg/*l* (Figures 4(D), 4(E) and 4(F)). In addition, the higher *K/S* value was observed in the 405-410 nm range.

SEM Analysis

The surface morphology of the cationized cotton fabric dyed with CFEE/AgNPs, CFME/AgNPs, and CFAE/AgNPs in both the absence and presence of a crosslinking agent were determined using SEM (Figure 5). Figures 5(A) and 5(B) show that the fabrics dyed with CFEE/AgNPs and CFME/AgNPs, respectively, were coated with uniformly dispersed spherical shape AgNPs, while the fabric dyed with CFAE/AgNPs exhibited mono dispersed spherical shape AgNPs (Figure 5(C)). In contrast, the SEM images of cationized fabric dyed in the presence of a crosslinking agent with CFEE/AgNPs and CFME/AgNPs in Figures 5(D) and 5(E) reveal a large quantity of spherical AgNPs in a clustered formation on the surface of the fibers. In addition, Figure 5(F) shows that the surface of the fabric dyed with CFAE/AgNPs exhibit spherically shaped AgNPs with a broad distribution of sizes.

EDX Analysis

The EDX spectra of fabrics dyed with CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs in the absence and presence of a crosslinking agent are presented in Figure 5(A-C) and Figure 5(D-F), respectively. The results that each figure exhibits a peak at ~3 KeV; this was attributed to the presence of AgNPs on the dyed fabric surface and also confirmed the success of in situ dyeing process.

XRD Analysis

XRD analysis was performed to study the crystal structure of the matrix and confirm the presence of AgNPs on the dyed cationized cotton fabrics, which may affect the properties and functionality of the fabric (Figure 6). In general, it was evident that the intensity of peaks in the cationized cotton fabrics was higher than the dyed fabrics, indicating that their crystallinity decreased in the presence of CF/AgNPs dyes. Figure 6(A) shows that the cationized cotton fabric exhibited sharp diffraction peaks at 2θ= 14.91 °, 16.60 °, 20.55 °, 22.921 °, and 34.41 ° which are characteristic of the crystalline structure of cellulose I [43]. However, the fabrics dyed with CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs did not only exhibit the peaks that are typical of cellulose I structure; they also exhibited an additional five, two, and one 2θ distinct diffraction peaks, respectively. These were attributed to the (111), (200), (220),

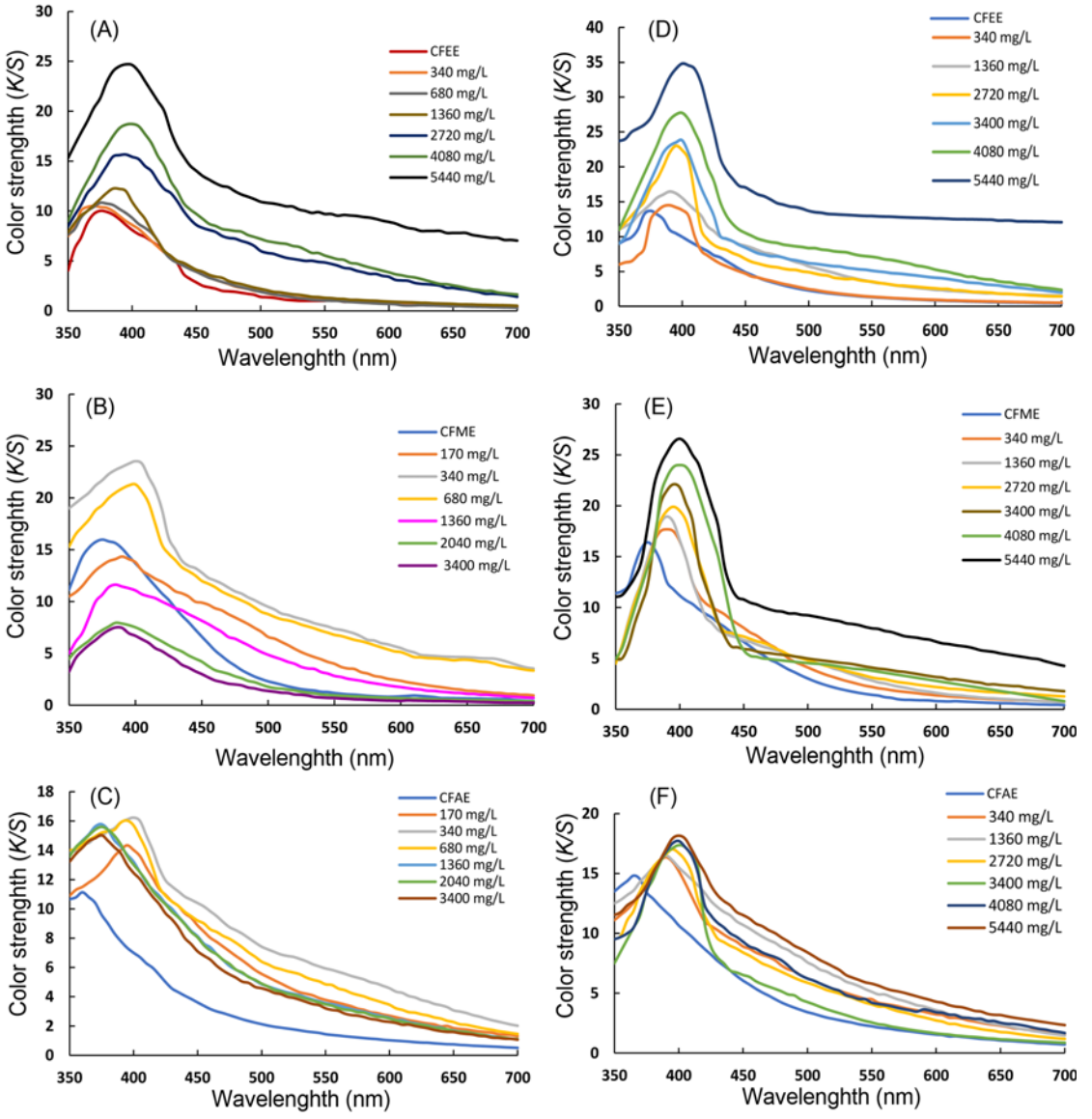


Figure 4. UV-Vis absorbance spectra of the dyed cationized cotton fabric with; (A) CFEE/AgNPs, (B) CFME/AgNPs, and (C) CFAE/AgNPs in the absence of crosslinking agent and (D) CFEE/AgNPs, (E) CFME/AgNPs, and (F) CFAE/AgNPs in the presence of crosslinking agent.

and (311) crystal planes of face-centered-cubic silver (FCC Ag).

The XRD analysis of the undyed and dyed cationized cotton fabrics in the presence of a crosslinking agent is presented in Figure 6(B). The undyed fabrics exhibited four sharp peaks that were characteristic of the crystalline form of cellulose I [47]. In contrast, the XRD pattern of the cotton fabrics dyed with CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs exhibited additional diffraction peaks that were distinct from the peaks associated with the crystalline structure of cellulose I.

Effects of Extraction Solvents on the Colorimetric Coordinates of Cotton Fabrics Dyed with CF Extracts

Table 4 illustrates the effect of the three solvents on the CF extracted dyes as well as the AgNPs dyes on the color strength values of the cationized cotton fabrics and their characteristic color values (L^*, a^*, b^*). Table 4 clearly shows that cationized cotton fabric dyed with CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs in the absence and presence of a crosslinking agent exhibited darker shades while also significantly improving the *K/S* values of the dyed fabrics. In particular, CFEE/AgNPs and CFME/AgNPs

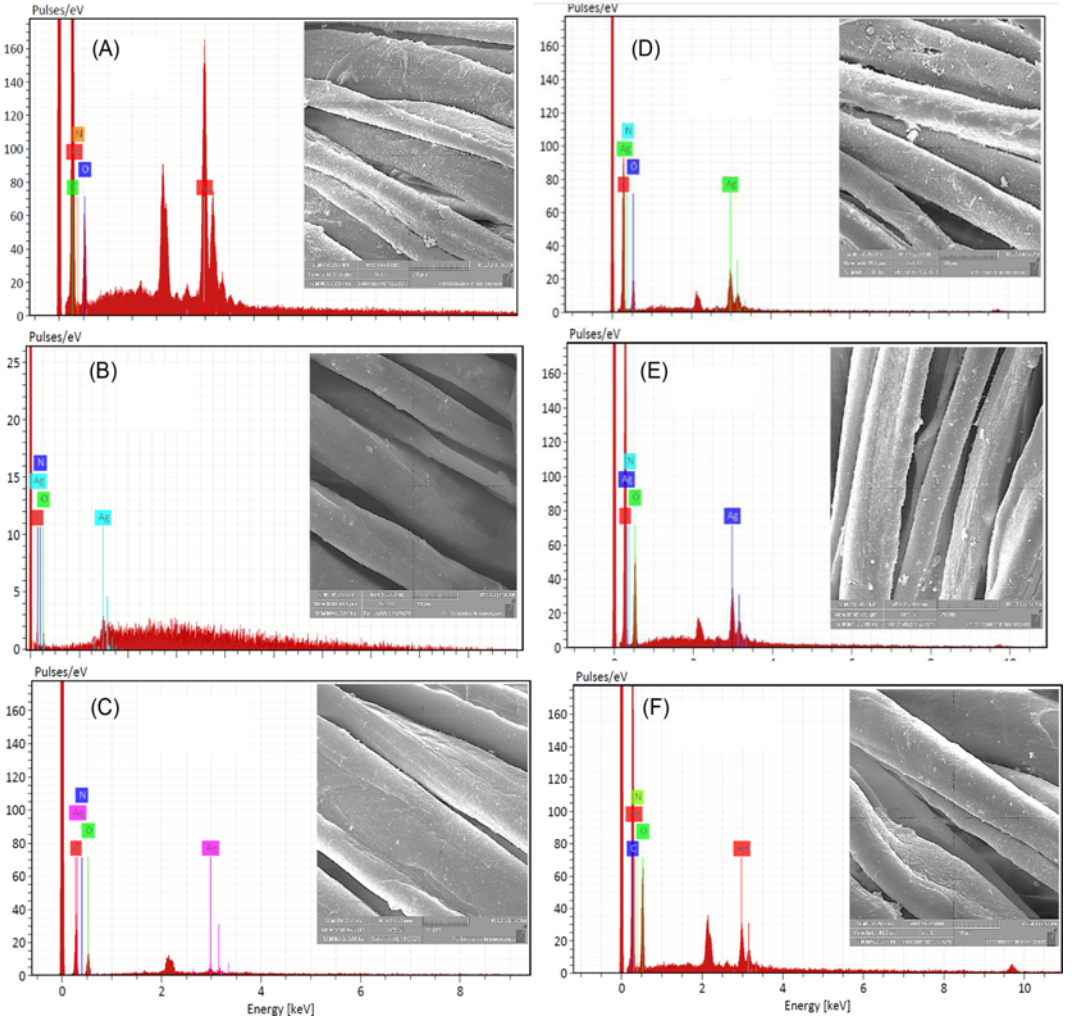


Figure 5. SEM and EDX of the dyed cationized cotton fabric with; (A) CFEE/AgNPs, (B) CFME/AgNPs, and (C) CFAE/AgNPs in the absence of crosslinking agent and (D) CFEE/AgNPs, E-CFME/AgNPs, and (F) CFAE/AgNPs in the presence of crosslinking agent.

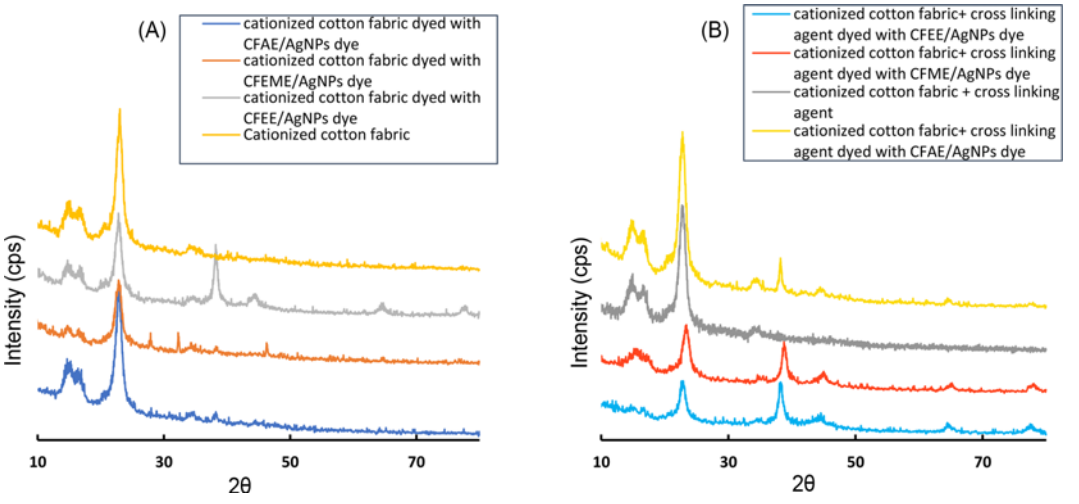


Figure 6. X-ray spectra of the undyed and dyd cationized cotton fabric with; (A) CFEE/AgNPs, CFME/AgNPs, and CFAE/AgNPs in the absence of crosslinking agent. (B) CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs in the presence of crosslinking agent.

Table 4. Colorimetric data of cotton fabric dyed with CFE dye in different solvent

Sample	L^*	a^*	b^*	*K/S*
Cationized cotton fabric dyed with:				
CFEE	67.33	5.16	31.77	10.01
CFEE/AgNPs	25.68	8.83	7.30	24.63
CFME	56.55	2.76	35.74	15.98
CFME/AgNPs	30.59	3.56	12.22	23.54
CFAE	49.53	6.87	24.92	11.13
CFAE/AgNPs	37.85	5.25	15.00	16.23
Cationized cotton fabric+crosslinking agent dyed with:				
CFEE	54.58	6.76	31.85	13.69
CFEE/AgNPs	21.99	9.13	3.10	34.80
CFME	53.12	8.94	36.93	16.41
CFME/AgNPs	25.16	4.77	4.78	26.58
CFAE	46.98	6.79	24.47	14.82
CFAE/AgNPs	33.28	6.11	12.19	18.15

shifted the color of the fabric to black and blackish brown shades on the red-yellow scale of the CIE color space. In contrast, CFEE and CFME dyes shifted the color coordinates toward the green region or zone of the color space [48]. In addition, all the dyed fabrics exhibited positive values for both a^* and b^*.

Color Fastness Properties

The results of the color fastness with respect to washing for each of the dyed cationized cotton fabrics are reported in Table 5. Each fabric was subjected to five cycles of washing. Cationized cotton fabrics dyed with CFEE, CFME, CFEE/AgNPs and CFME/AgNPs achieved a good rating. However, cationized cotton fabrics dyed with CFAE and CFAE/AgNPs only managed to obtain a moderate rating. This may be due to the presence of ionic linkages and non-polar van der Waals forces of attraction between the more bioactive phenolic coloring components present in the hydroalcoholic CF extracts and the cationized cotton fabrics. In addition, cationized cotton fabrics dyed with CFEE, CFME, CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs in the presence of a crosslinking agent obtained good ratings, while the fabric dyed with CFAE dye only achieved a moderate rating. In general, the dyed cationized cotton fabrics exhibited moderate to good ratings for the color fastness index with respect to laundry washing.

Multifunctional Finishing Properties of CF Extracted Dyes

UV Protection Properties

The UPF of cationized cotton fabrics dyed with CF extracted dyes and with CF extracted/AgNPs dyes in the presence and absence of a crosslinking agent was represented in Table 6. Blank cotton fabric was used as a control. The results indicate that the undyed cationized cotton fabric in the presence and absence of a crosslinking agent show lower UPF values. However, the fabrics dyed with CF extracts and CF extracted/AgNPs dyes exhibited increased UPF levels; this imparts the dyed fabrics with greater UV protection levels and increases the UV-blocking activity of the cotton fabrics. This may be due to the phytochemical composition

Table 5. Color fastness properties of cotton fabric dyed with CF extracted dyes in different solvent

Sample	Washing fastness durability														
	First cycle			Second cycle			Third cycle			Fourth cycle			Fifth cycle		
	Alt*	SC*	SW*	Alt*	SC*	SW*	Alt*	SC*	SW*	Alt*	SC*	SW*	Alt*	SC*	SW*
Cationized cotton fabric dyed with:															
CFEE	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
CFEE/AgNPs	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
CFME	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
CFME/AgNPs	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
CFAE	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4
CFAE/AgNPs	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	4	3-4	3-4	3-4	3-4	3-4
Cationized cotton fabric+crosslinking agent dyed with:															
CFEE	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
CFEE/AgNPs	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
CFME	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
CFME/AgNPs	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4
CFAE	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4
CFAE/AgNPs	4	4	4	4	4	4	4	4	4	4	4	4	4	4	4

Alt*: color change, SC*: staining on cotton and SW*: staining on wool.

Table 6. UV protection of cationized cotton fabric dyed with CF extracted dyes in different solvent

Sample	Conc.of $AgNO_3$ (mg/*l*)	Methanol extract	Ethanol extract	Aqueous extract
		UPF	UPF	UPF
Blank cotton fabric	0	2.80	2.80	2.80
Cationized cotton fabric	0	2.41	2.41	2.41
Cationized cotton fabric dyed with:				
CF extract	0	49.40	20.4	25.8
CF extract/AgNPs	170	57.54	20.8	27.2
	340	101.36	21.6	26.3
	680	38.21	22.0	22.4
	1360	31.21	22.4	28.7
	2040	28.21	21.8	23.6
	2720	27.65	22.7	29.5
	3400	24.03	23. 6	25.0
	4080	-	24.4	-
	5440	-	41.5	-
Cattionized cotton fabric+crosslinking agent	0	5.6	5.6	5.6
Cationized cotton fabric+crosslinking agent dyed with:				
CF extract	0	23.8	27.3	89.0
CFE/AgNPs	340	22.4	23.4	113.2
	1360	30.4	23.8	97.1
	2720	50.3	77.3	99.2
	3400	55.6	69.7	79.4
	4080	59.8	32.5	71.3
	5440	69.6	52.7	207.50

of the dyes extracted by each solvent, which contain phenolic and flavonoid compounds that exhibit free radical scavenging activity, which confers UV-blocking properties [49,50]. Cationized cotton fabrics dyed with CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs exhibited higher UPF values than CFEE, CFME and CFAE dyed fabrics due to the larger refractive index of AgNPs dyes, which leads to more robust UV scattering.

In addition, the UPF values of all cationized cotton fabrics dyed in presence of a crosslinking agent increased as the $AgNO_3$ concentration was increased. Hence, the increased deposition of AgNPs is associated with markedly higher UV absorption as a result of the darkened color of the dyed fabric, which increases the efficiency of UV scattering due to the large refractive index of AgNPs. Notable, cationized cotton fabric dyed with CFAE and CFAE/AgNPs exhibited the greatest UPF values. This may be due to the phytochemical composition of the aqueous extracted dyes, which is composed of phenolic acid, some flavonoids and other components, which could be very effective at scattering UV radiation. Additionally, these results confirm the presence of material with UV blocking properties in the cotton fabrics dyed with CF extracted dyes regardless of solvent. These are very efficient at protecting human skin against harmful UV radiation, and can thus be widely used for various medical applications [2,51].

Antimicrobial Activity

There has been an increasing demand for cellulosic fabrics with antimicrobial properties; these can be widely applied to the biomedical and health care sectors [52]. The antimicrobial activity of a blank fabric and the fabrics dyed with CF extracted dyes were evaluated against the five pathogenic microorganisms presented in Table 7. These pathogenic microorganisms were selected because they are the most common types of microbial infection in hospitals and they resist most antibiotics. As expected, the blank cotton fabric does not possess any antimicrobial activity. However, undyed cationized cotton fabric exhibited a low degree of antimicrobial activity as well as moderate resistance against fungal activity. Cationized cotton fabrics dyed with CFEE and CFME exhibited much higher antibacterial activities than fabrics dyed with CFAE due to the higher quantities of apigenin and its derivatives, as well as other polyphenolic bioactive compounds in the extracted dyes, which increase the antibacterial properties of the dyed fabric.

In general, cationized cotton fabrics dyed with CFEE/AgNPs, CFME/AgNPs, and CFAE/AgNPs in the absence or presence of a crosslinking agent were resistance to a wide

Table 7. Reduction percent of the pathogenic microorganisms of dyed cotton fabrics with CF extracted dyes in different solvent

Sample	Pathogenic microorganism (reduction %)				
	B. subtilis	*S. aureus*	*E. coli*	*P. aeruginosa*	*C. albicans*
Blank cotton fabric	0	0	0	0	0
Cationized cotton fabric	0	17.91	48.87	0	66.84
Cationized cotton fabric dyed with :					
CFEE	24.89	71.65	77.37	94.60	74.11
CFEE/AgNPs	78.61	92.91	79.45	96.40	77.65
CFME	66.00	57.54	40.19	75.43	65.10
CFME/AgNPs	78.71	88.76	87.78	88.11	81.86
CFAE	43.15	38.55	65.28	41.73	41.21
CFAE/AgNPs	50.21	40.72	67.18	45.61	65.88
Cationized cotton fabric+crosslinking agent	8.51	49.83	34.78	63.95	76.81
Cationized cotton fabric+crosslinking agent dyed with:					
CFEE	10.21	19.37	27.12	37.62	25.15
CFEE/AgNPs	44.15	97.01	78.33	91.01	83.35
CFME	58.10	29.34	9.32	42.32	32.80
CFME/AgNPs	60.32	80.99	33.91	55.81	83.39
CFAE	81.38	30.10	17.21	32.21	75.82
CFAE/AgNPs	85.51	78.97	31.31	41.12	48.36

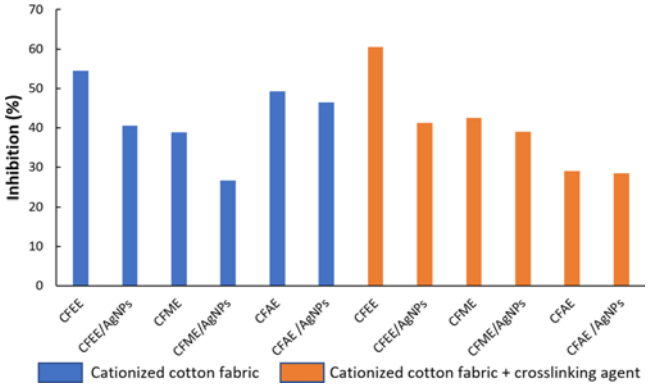


Figure 7. Antioxidant activity of dyed cotton fabrics.

range of pathogenic microorganisms compared to those dyed with CFEE, CFME and CFAE. However, this was dependent upon the solvent used for dye extraction, the concentration of $AgNO_3$, and the bacterial strains. Several hypothetical mechanisms can explain the antimicrobial activities of dyed fabric, as follows:

1) The interactions between the -OH phenolic groups of the CF dye components and the $-NH_2$ or -COOH groups of the bacterial cell wall proteins can form hydrogen bonds, which can consequently deactivate or damage the cell wall proteins [53].

2) Several mechanisms have been reported concerning the antimicrobial properties of AgNPs, but the specifics of these mechanisms were unclear. Some hypothesized mechanisms are as follows:

a. AgNPs attached to the surficial biomolecules of the bacterial cell can alter its properties and disturb the negatively charged structural lipopolysaccharide. They can also accumulate inside the cell membrane by forming pits, incrementing the permeability of the membrane and causing cell death.

b. AgNPs can enter the cells of the microorganism, causing damage to the cell's DNA.

c. The dissolution of AgNPs into Ag^+ ions can bind with negatively charged species in the cell membrane of the microbe by electrostatic attraction, and inhibiting the growth of colonies [2,54].

Antioxidant Activity

Natural polyphenols and flavonoid compounds are considered to be major natural antioxidants, and play a vital role in human health due to the free radical scavenging activity displayed by hydroxyl groups. The production of natural bio-colorants and bioactive textile fabrics via the simultaneous dyeing process and the green chemistry approach can be significant in terms of its application to various medical fields [55]. This may be due to the trapping of free oxygen radical species, which prevents cell damage and growth of new cells in the skin [56]. Figure 7 reveals that the cationized cotton fabrics dyed with CFEE, CFME and CFAE exhibited higher antioxidant activities depending upon the phytochemical constituent of CF dye extracts. Hence, the antioxidant activity exhibited by these fabrics may be due to the high average concentration of phenolics

and flavonoid compounds present on the surface of dyed fabrics. In contrast, the cationized cotton fabrics dyed with CFEE/AgNPs, CFME/AgNPs and CFAE/AgNPs exhibited moderate antioxidant activities, which may be attributed to the consumption of a fraction of the phenolic compounds (in particular, the -OH groups) during the reduction and chelating of Ag^{+} to AgNPs during the dyeing process. The major antioxidative activity of CF extracted dyes can be divided into phenolic acid compounds (gallic acid, caffeic acid, coumaric acid, ferulic acid, etc.) and flavonoids, which include catechin, apigenin, quercetin, leuteolin, etc). Phenolic acids serve as antioxidants by potentially trapping free radicals, while flavonoids grant the dyes the ability to better scavenge free radicals or chelate metals [57,58].

Conclusion

This study has demonstrated the efficiency of bioactive compounds in dyes extracted from CFs using different solvents, as well as their potential for use as multifunctional dyes in the one pot approach to the green synthesis of AgNPs when dyeing cationized cotton fabrics. This introduces a facile route for the robust dyeing of cotton fabrics with multifunctional properties. Our results showed that the 80 % ethanol and 80 % methanol extracts contain more TPC (1523.80 μg GAE/m*l*, and 1398.62 μg GAE/m*l*, respectively) and TFC (845.91 μg CE/m*l* and 678.75 μg CE/m*l*, respectively) than the aqueous extracts (1018.00 μg GAE/m*l* and 444.11 μg CE/m*l*) after the extraction conditions were optimized. In addition, the bioactive components of the methanol and ethanol CF extracts exhibited higher biological activity than the aqueous extract. However, all of them could be potentially used as natural dyes for the dyeing of cationized cotton fabrics. A diverse range of colors, including greenish yellow, golden yellow and yellowish brown was obtained; they exhibited moderate to good washing fastness, excellent antioxidant activity, good UV protection but inadequate antimicrobial properties. The application of CF extracted dyes successfully played a dual role as bio-reducing and stabilizing agents for the green synthesis of AgNPs dyes, and the one pot approach to dyeing the cotton fabric resulted in a diverse range of dark colors, including black, blackish brown and dark browns. The fabrics dyed with these CF extracted AgNPs dyes exhibited good washing fastness and excellent antibacterial activities against a variety of pathogenic microorganisms and as well as excellent fungal resistance towards *C. albicans*. Furthermore, each of the dyed fabrics exhibited better UV protection in the presence of AgNPs dyes as well as lower antioxidant activities than cationized cotton fabric dyed with CFEE, CFME and CFAE. Hence, we believe that the development of dyed cotton fabrics with multifunctional finishing using value-added dyes are essential for the biomedical textile sector.

Conflicts of Interest

Authors declare that they have no conflict of interest.

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