Functional and antimicrobial properties of herbal nanocomposites from *Piper betle* plant leaves for enhanced cotton fabrics

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Abstract In the current times, the application of nanoparticles in the textile industry has become increasingly high due to the possibility of having anticipated properties, such as captivating colors, superior stability, antibacterial activity, and high-end UV-protection to the fabrics. In this study, natural herbal nanoparticles of different sizes were prepared from shade-dried leaves of *Piper betle* employing ball milling technique. Going forward, structural, morphological, UV-protective, and antibacterial properties of herbal nanocomposites coated on fabrics were thoroughly analyzed and interrelated with uncoated fabrics. Herbal nanoparticles were amalgamated with chitosan to make nanocomposites and are coated on cotton fabrics with the help of the pad-dry cure method. The analysis done to study physical properties of the coated fabrics, such as air permeability, crease recovery angle, tensile strength, tearing strength, thickness, and bursting strength, explicitly showed that coated fabrics have better functional properties as compared to uncoated fabrics. Along the same lines, herbal nanoparticles reflected good antibacterial and UV-absorption properties as compared to uncoated and chitosan-coated fabrics. Comprehension of functional properties revealed that herbal nanoparticle-coated fabrics highlights the potential applications of *Piper betle* nanoparticles in protective clothing.

Keywords *Piper betle*, Herbal nanoparticles, Nanocomposite, UV-protection, Chitosan, Antibacterial activity

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Introduction

Nanoparticles play a critical yet unique role in an array of fields due to their unmatched properties and applications.[1] A textile is an impeccable growth medium for microbial growth as it is rich in organic compounds that are appropriate for biofilm formation and sweat absorption, This, in turn, is responsible for providing moisture conditions and causing infections to humans.[2–4] Commonly, *Staphylococcus aureus* (*S. aureus*) and *Staphylococcus epidermidis* (*S. epidermidis*) lead to skin infections like boils, impetigo and cellulitis, and furuncle.[5,6] In a textile, microorganisms bring about negative effects such as dreadful odor, discoloration and reduced texture efficiency.[7] The aforementioned issue is conventionally combated by using an antibacterial finishing process with the help of aldehydes, halogens, quaternary ammonium compounds, and amines.[8]

Chemical antibacterial finishing results in the best control of microbial growth, but it also accompanies some drawbacks as well, such as being toxic, nonbiodegradable, not eco-friendly, and cost-effective. Hence, it is indispensable to change reliable, nontoxic, nonallergic, eco-friendly materials for textiles with antibacterial finishing. The

aforementioned factors are possible by making use of biological materials, particularly herbal plants. Additionally, the utilization of plant extracts and bioactive compounds encompasses a range of antibiotic properties, which is conventionally utilized in therapeutic treatments.[9,10] Moreover, herbal molecules are secure as they can easily overcome the resistance generated by the pathogenic microbes. This is possible because they are in collective form that contains more than one molecule in the protoplasm of the plant cell.[11]

In this study, we have selected one of the herbal plants, i.e., *Piper betle* that belongs to *Piperaceae* family. It has good antibacterial activity against *S. aureus, Proteus vulgaris, Escherichia coli* (*E. coli*), and *Pseudomonas aeruginosa*. Typically, the antibacterial activity occurs because of the presence of sterol in leaf extract of *Piper betle*. The mode of action is through surface interaction with the bacterial cell wall and cell membrane and alters the primary structure of the cell wall, leading to the generation of pores and worsening of both Gram-positive and negative bacteria.[12–14] *Piper betle* encompasses phenol components, like ally pyrocatechol, chavicol, chavibetol, carvacrol, eugenol, and safrole.[12,14,15] Ally pyrocatechol and chavibetol confer radio-protective activity with the process inhibition through the radiation-induced lipid peroxidation mechanism.[12,16] When it is used for textile applications, such compounds can provide better antibacterial activity and UV-protection.

In textiles, antibacterial agents are coated on fabrics using chemical agents for binding. Bionanocomposite materials are currently used in material science and nanoscience fields. Bionanocomposites consist of polymeric compounds with materials on the nanoscale.[17,18] Chitosan is a glucosamine biopolymer with better reactive amino groups, antimicrobial, biocompatibility, exceptional film-forming nature, and nontoxicity.[18–22] In addition, it has this positively charged polymer bond with negatively charged surface, mucoadhesive, and wound-healing properties which are attractive for biomedical applications.[23–25]

In the current study, *Piper betle* nanoparticles are produced by using shade-dried plant leaves with the help of ball milling. As prepared, *Piper betle* nanoparticles are preliminarily observed to take up qualitative studies for surface morphology, size, crystallinity, and purity. Moving ahead, the synthesized herbal nanoparticles are blended with chitosan to prepare a polymeric nanocomposite for coating on fabric. The coated fabrics are thereafter subjected to UV-protection and antibacterial investigation. Also, functional studies such as strength (tensile and tearing) properties, air permeability, and crease recovery are tested.

Materials and methods

Plant leaf sample collection

Fresh, healthy, and mature *Piper betle* leaves without disease symptoms were collected from places in Namakkal, Tamil Nadu, India. Then, the leaves were thoroughly washed with running tap water, including double-distilled (DD) water, for several times to alleviate all the dust particles. Moving ahead, the leaves were then air-dried under shade at room temperature for around 2 weeks.

Preparation of herbal nanoparticles

The 20 g of shade-dried leaves was primarily ground via top-down approach with the help of the mixer grinder to prepare the coarse powder. In order to obtain the fine powder, the coarse powder was milled for 1 h with the help of 20-mm-sized ball (Zirconia) by making use of ball mill (PM100; Retsch, Germany). Along with this, the obtained fine powder was (nearly 7 gm) distributed into three equal parts. Then, these equal fine powder sections were again milled (10 mm balls: 300 rpm) in a ball mill at distinct milling periods: 5, 10, and 15 h (hereafter termed, respectively, as samples PBNp1, PBNp2, and PBNp3).[26–29]

Figure 1 schematically represents the preparation of herbal nanoparticles from *Piper betle* plant leaves. Thereafter, the nanoparticles obtained from the three collected samples (PBNp1, PBNp2, and PBNp3) were characterized carefully and were further utilized for coating on fabrics. This was followed by the analysis of their functional properties.

Characterization

The phase finding of *Piper betle* nanopowders was performed with the help of X-ray diffraction method utilizing the (Philips XRD; X' Pert Pro; PANalytical, Netherlands) X pert MPD powder diffractometer. The powder XRD was obtained in the 2θ range from $10°$ to $80°$ in a step-scan mode with 2θ step of $0.02°$. When bombarded with high-energy X-rays for elemental analysis, the obtained leaf nanoparticles were excited.

Further, the synthesized leaf nanopowders were carefully monitored on a periodic basis in a UV–visible (UV–Vis) spectrophotometer (Cary 8454, Agilent technologies, Singapore) functioned from the UV to near-infrared (NIR) (180–800 nm) spectral regions at a step size of 5 Å. Thereafter, a volume of 0.1 mL of the sample was diluted in a cuvette with 2 mL of deionized water. At a resolution of 1 nm, the UV–Vis spectra of the resulting diluents were analyzed as a function of reaction time and biomaterial dosage. Using dynamic light scattering (DLS) technique, the size of the particles was analyzed with the help of a submicrometer particle size analyzer (Nanophox, Sympatec, Clausthal-Zellerfeld, Germany). The particle size of all the obtained herbal nanopowders samples was measured at a scattering angle of $90°$ in the range of 1–1000 nm. The prepared herbal nanoparticles were thereafter taken to the scanning electron microscope equipped with energy-dispersive X-ray analysis (SEM–
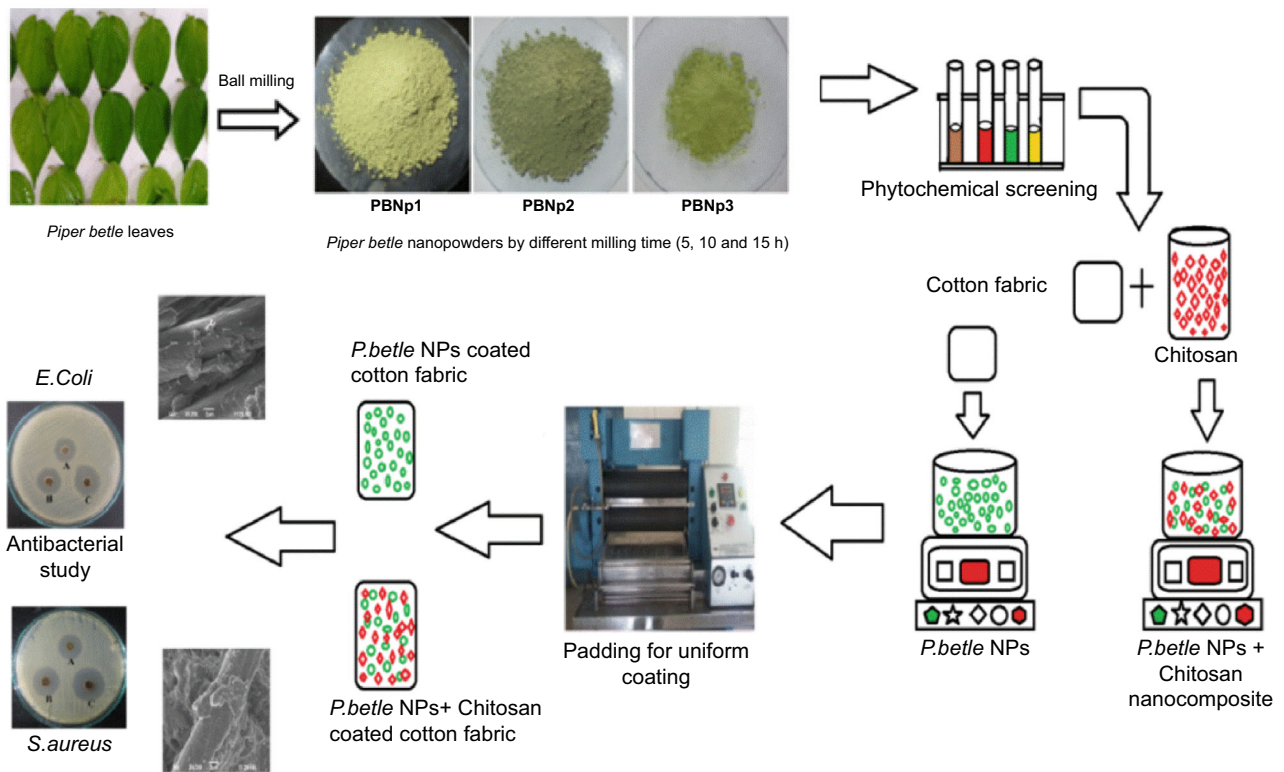
Fig. 1: Schematic representation of the preparation of herbal nanoparticles from *Piper betle* plant leaves

EDX; JSM 6360; JEOL, Japan) to determine the morphology, microstructure, and elemental composition of the collected samples.

Preparation of herbal nanoparticles chitosan composite

The obtained herbal nanoparticles (1 g) were mixed in 100 mL of double-distilled water, including 1% chitosan (from shrimp shells, ≥ 75% deacetylation, Himedia, India), dissolved in 1% acetic acid. The solution was kept under stirring at 60°C until a fine homogenous suspension formed. Thereafter, the obtained suspension was kept overnight to alleviate all air bubbles. Along with that, the solution was sonicated for 30 min before use.[6]

Next, the bleached and mercerized woven cotton fabric (100%, mass 138.84 g m^{-2}, 116 ends per inch 84 picks per inch) was pretreated as a substrate for coating. Then, the cleaned fabrics were dried in an oven at 50°C for around 5 min. It is imperative to add chitosan in order to make the herbal nanoparticles stable in an aqueous solution for a longer period of time. A bleached (60 × 60 cm^2) cotton fabric was then individually immersed four times in a homogeneous solution containing chitosan sol and *Piper betle* chitosan solution (nanocomposite finishing). Then, the obtained solution was passed through a padding mangle at a rate of 35 rpm for around 5 min with the help of a DC motor to collect the uniformly coated fabric. Thereafter, it was dried at 80°C for 10 min.[6,30]

Ultraviolet protection on coated fabrics

Ultraviolet transmission spectra (Lambda 35; PerkinElmer, USA) with the wavelength ranging from 280 to 400 nm were used to test the UV-blocking properties of the fabric samples. The total percentage of UV blocking was measured with respect to ASTM D6603 standard.[31] Below is the value of the UV-protection factor (UPF) that was obtained with the help of the following relation:

$$\text{UPF} = \frac{\sum_{290}^{400} E(\lambda) \times S(\lambda) \times \Delta\lambda}{\sum_{290}^{400} E(\lambda) \times S(\lambda) \times T(\lambda) \times \Delta\lambda} \tag{1}$$

where $S(\lambda)$ stands for the solar spectral irradiance (W m^{-2} nm^{-1}), $E(\lambda)$ stands for the relative erythemal spectral effectiveness, $T(\lambda)$ stands for the spectral transmittance, and $\Delta(\lambda)$ stands the wavelength interval.

Physical properties

Further, the thickness of the coated and uncoated fabric samples was obtained with the help of fabric thickness tester with respect to ASTM D5729-97 guidelines, ranging from 0 to 10 mm. The tensile strength of the uncoated fabrics and coated fabrics was evaluated by strip method and tensile testing machine (E091; Eureka, India) with respect to ASTM D5035-95 standard.[32,33] Apparently, the testing was auctioned utilizing warp and weft yarns of the prepared fabric samples. Also, the crease recovery analyzer (EC-41; Eureka, India) was used to calculate the crease recovery angle in accordance with AATCC 66-1998 standard with a scale ranging from 20° to 80° at an applied creasing load of 300 g for 2 min.[34] The air permeability values of coated and uncoated fabrics were obtained as per IS: 11056-1984 and DIN 53887 standards with the flow rate of 100 Pa for passing the air perpendicularly utilizing an air permeability tester (M021A; Premier, India).[35]

Antibacterial activity

Collection of bacterial cultures and preparation

The cultures of Gram-positive *S. aureus* (ATCC 6538P) and Gram-negative *E. coli* (ATCC 9677) were obtained from the National Collection of Industrial Microorganisms (NCIM), National Chemical Laboratory, Pune, India. Bacterial inoculum was prepared by carefully inoculating a loop full of test organisms into a nutrient broth and then incubating at 37°C for 5–8 h until a moderate turbidity was developed. Going forward, a loop of culture was swabbed on the Mueller–Hinton agar media (Himedia, Mumbai).

Agar well diffusion method

The qualitative antibacterial evaluation was performed for *E. coli* and *S. aureus* using agar well diffusion method. A fine well was made in the solidified and inoculated culture medium (Mueller–Hinton agar) utilizing a sterile cork borer. The diameter of 7 mm was maintained on each zone as in all plates. All the wells were loaded with distinct mass concentrations such as 25, 50, and 100 mg mL^{-1} of each prepared PBNp1, PBNp2, and PBNp3 herbal nanoparticles. The plates were then incubated at 37°C for approximately 24 h. After completion of the incubation period, the diameters of the inhibition zones were calculated using a millimeter ruler.

Agar disk diffusion method

According to standard size (each piece 10 mm in size), the uncoated cotton fabrics (UC-CF) and poly-mer/nanocomposites-coated (CF-Chi and CF-PBNp-Chi) fabrics were divided into different sample pieces. Then, the prepared specimen was carefully pressed in a transverse manner across the inoculums of streaks. This was done to ensure close contact with the agar surface. In addition, the plates were incubated for 18–24 h at 37°C. Then, the aforementioned process was followed to perform the antibacterial assessments of UC-CF, CF-Chi, and CF-PBNp-Chi fabrics against *E. coli* and *S. aureus*.

Percentage reduction test

In this test, the uncoated (UC-CF) and poly-mer/nanocomposites-coated fabrics (CF-Chi and CF-PBNp-Chi) were cut into round-shaped pieces (10 mm) as recommended by the American Association of Textile Chemists and Colonists (AATCC 100). The obtained test samples were then properly soaked into sterile AATCC bacteriostasis broth. After that, a loop full of test organisms was inoculated. The obtained test samples were incubated at 37°C for around 18–24 h. Then, serial dilutions from 10^{-1} to 10^{-7} were made for all the prepared samples. By using the spread plate method, 0.1 mL of sample from each dilution was plated on the sterile AATCC bacteriostasis agar plates and then incubated at 37°C for 24 h. Upon completion of the incubation period, the final concentration of colonies (B) in control and the obtained test samples were measured using viable cell count method. As the untreated fabric (control) contained no bactericidal activity, the final number of surviving cells would be greater than the initial cell concentration (A), which was calculated through a viable cell count method.[6] The percentage of bacterial reduction was calculated with the help of the following formula:

$$\% \text{ Bacterial Reduction} = [(A - B)/A] \times 100 \qquad (2)$$

where A stands for the initial number of cells and B for the final number of cells.

Wash durability of coated fabrics

The IS: 687-1979 standard was utilized to determine the wash durability of CF-Chi and CF-PBNp-Chi fabrics.[36] This test was performed using a neutral soap at 40 ± 2°C for around 30 min. The test fabrics were dried out to assess the control for antibacterial activity with the help of AATCC 100 procedure, which involved up to 15 laundering cycles.[37]

Results and discussion

Characterization of P. betle nanoparticles

The shade-dried *Piper betle* herbal nanoparticles were prepared by ball milling. The obtained nanoparticles were milled at different milling periods (5, 10, and 15 h) at 300 rpm. Then, the nanoparticles were characterized by using different characterization techniques. The XRD pattern of the prepared nanoparticles, namely PBNp1, PBNp2, and PBNp3, is depicted in Fig. 2A. The obtained results ensure that there are no diffraction peaks at 2θ values in the range of 20°–30°. The experimental results explicitly confirm that different parameters of samples depict amorphous nature with no crystalline peaks.

Figure 2B shows the UV-absorption spectra of herbal nanoparticles synthesized from *Piper betle* plant leaves. In this analysis, all three rotation periods of *Piper betle* possess the same UV-absorbance region that ranges from 280 to 284 nm. Such absorbance (280–282 nm) in the UV region allows utilization of the herbal PBNp nanoparticles for UV-protection applications. Figure 2C shows the particle size analysis of PBNp1, PBNp2, and PBNp3. It is evident from Fig. 2C that the average particle size of PBNp1, PBNp2, and PBNp3 samples is 81, 55, and 26 nm, respectively. These are found to be decreased in sizes from 25 to 30 nm with an increase in rotation.

The surface morphological properties of PBNps are evident from the SEM images (Fig. 2D). The topography of the SEM image shows clear signs of herbal PBNp nanoparticles that appear to be agglomerated with cluster like structure. As expected, the elemental analyses confirm the presence of identical elements across all the collected samples (Table 1). In this study, the high radiation at high energies for O, Na including at low energies for Mg, Si, Cl, K, and Ca is seen for PBNp nanoparticles (Fig. 2D).

The qualitative evaluation of the antibacterial activity of the herbal nanoparticles (PBNp1, PBNp2, and PBNp3) possesses intriguing observations, which is quite evident from Fig. 2E. The zone of inhibition at diverse concentrations of different ball milling times (PBNp1, PBNp2, and PBNp3), namely 25, 50, and 100 mg mL^{-1} of PBNp nanoparticles, is shown in Fig. 2E and later concluded in Table 1.

The highest formation of zone of inhibition for *E. coli* (27.24 ± 0.12 mm) and *S. aureus* (31.20 ± 0.13 mm) was observed for the sample PBNp3. This indicates a higher time period of milling (15 h) of the plant leaf powders. At this increased milling time, the size of the nanoparticles reflected is only 26 nm, which is very low compared to other milling times. The smaller particles are observed to possess the formation of maximum zone of inhibition as compared to the larger particles. The larger surface area of smaller nanoparticles aids in seamless penetration and denaturing of the bacterial cell wall. This, in turn, restrains the DNA replications[38–40] and thereby leads to formation of wider zone of inhibition than the small surface area of larger nanoparticles.[26] The current studies ensure that the surface area of nanoparticles has an impact on antibacterial property of PBNps. By carefully studying the characterization outcomes of all three collected samples of PBNp, PBNp3 is opted for coating process and therefore indicated as PBNp.

Characterization of uncoated and coated fabrics

Figure 3a illustrates the surface morphological properties and EDX analysis of uncoated cotton fabrics (UC-CF). On the fabric surface [Fig. 3b (i, ii)], the SEM image of the chitosan-coated cotton fabrics (CF-Chi) elucidates cluster-type polymer structures prior to wash. Figure 3c (i, ii) reveals the SEM image and EDX analysis of the CF-PBNps-Chi nanocomposites which are clearly immersed on the fabric surface area.

The chitosan polymer is sturdily adhered to the surface of the fabrics even after 5th and 10th washes [Figs. 4a(i) and 5a(i)]. Thereafter, the wash durability (fastness) of CF-Chi fabric surface is further ensured with the help of EDX measurements [Figs. 4a(ii) and 5a(ii)] carried out after 5th and 10th washes. Figure 4c elucidates the even dispersion of PBNp-Chi nanocomposite particles.

Moreover, elemental composition analysis on coated fabrics prior to wash is also reflected. The presence of PBNp-Chi nanocomposite particles and their wash durability after 5th and 10th washes are proven through SEM and EDX analysis, respectively, in Figs. 4b(i, ii) and 5b(i, ii).

The aforementioned observation clearly shows that the CF-Chi and CF-PBNp-Chi fabrics possess high adhesion on the fabrics even when they undergo multiple washes. However, the percentage of nanoparticles coated on the fabric gets significantly reduced. This implies that more nanoparticles are evident after 5th wash than after 10th wash. This suggests that more washes lead to alleviation of particles coating from the fabric surface. This is again fostered by the alternations in the calculated thickness of the UC-CF, CF-Chi, and CF-PBNp-Chi fabrics before and after wash (Table 2). Nevertheless, concentration of nanoparticles present on fabric surface is enough to regain their functional properties, even up to a maximum usage life of the fabrics, i.e., 10th wash. In many other applications like wound dressing and biomedical, antibacterial fabrics are hired for one time use only.

Additionally, the coating thickness of nanoparticles on fabric clearly shows the air permeability of the fabrics. Air permeability test on UC-CF and CF-Chi, including CF-PBNp-Chi, fabrics observed that permeability of the fabric shows a decrease of 85% in air permeability as compared to those of UC-CF fabric and CF-Chi fabric that reflected a 53% reduction. It was concluded that the low air permeability in CF-
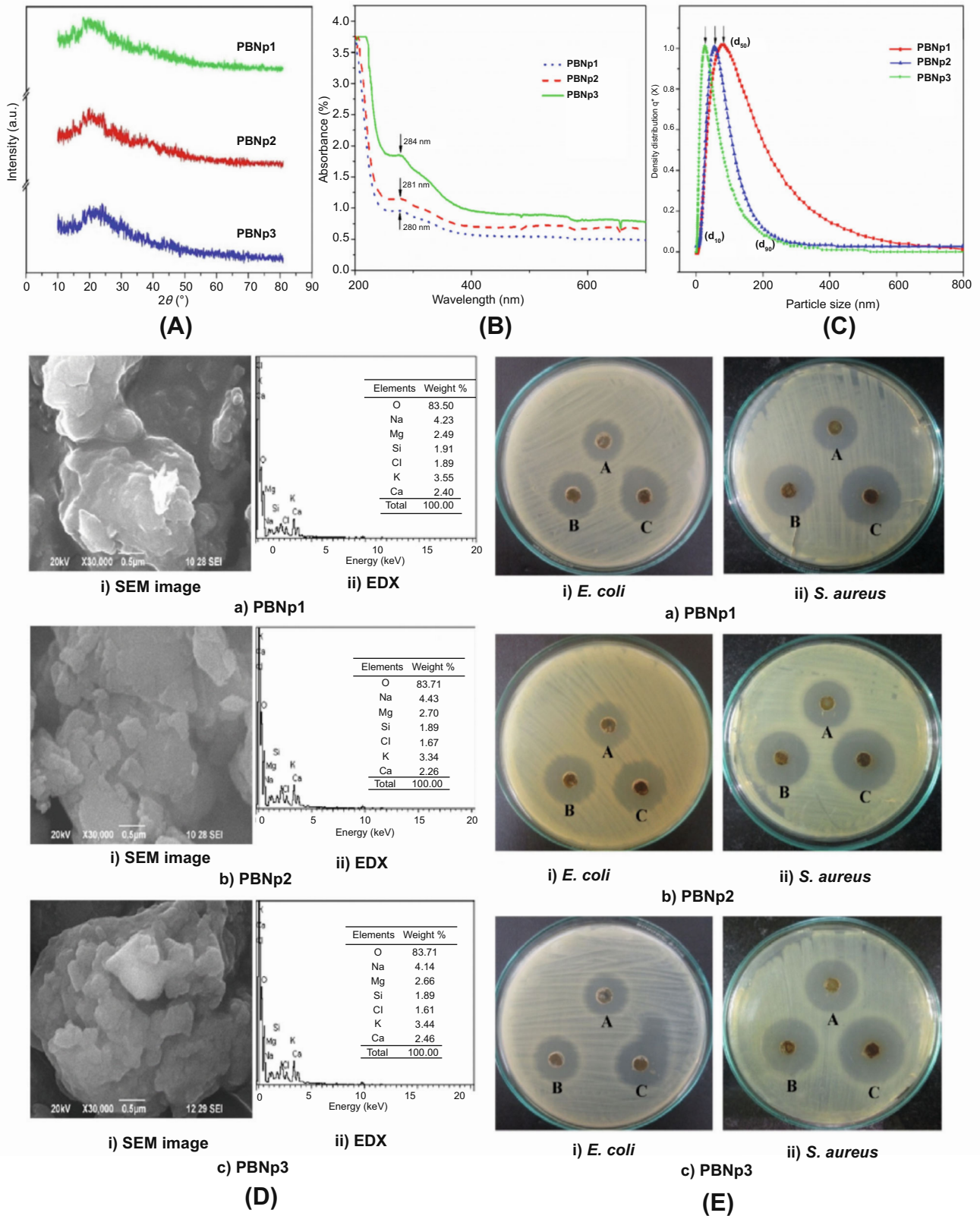
Fig. 2: XRD (A), UV–Vis spectrum (B), particle size distribution (C), SEM/EDX (D), and antibacterial activity (E) of three different milling periods (PBNp1, PBNp2, and PBNp3) for PBNp nanoparticles

Table 1: Antibacterial activity of the *Piper betle* nanoparticles by agar well diffusion assay

Herbal nanoparticles	Test organisms	Concentrations of *Piper betle* nanoparticles [zone of inhibition (mm)]		
		25 mg mL^{-1}	50 mg mL^{-1}	100 mg mL^{-1}
PBNp1	*E. coli*	18.52 ± 0.43	21.40 ± 0.12	23.00 ± 0.08
	S. aureus	20.08 ± 0.42	23.03 ± 0.28	27.20 ± 0.21
PBNp2	*E. coli*	19.74 ± 0.02	22.68 ± 0.32	25.56 ± 0.41
	S. aureus	22.20 ± 0.23	25.20 ± 0.03	28.38 ± 0.06
PBNp3	*E. coli*	20.14 ± 0.05	24.50 ± 0.07	27.24 ± 0.12
	S. aureus	23.80 ± 0.06	26.07 ± 0.05	31.20 ± 0.13

PBNp fabrics is due to the absorption of composite materials in between the fibers of the fabric. The air permeability allied to CF-PBNp-Chi fabric after 5th and 10th washes improves to 65% and 72%, respectively. Also, the chitosan-coated fabric reflected an upsurge of 60% and 71%, respectively. The increase in air permeability after 5th and 10th washes is due to alleviation of the coatings from the fabric surface.

The mechanical properties of the CF, CF-Chi, and CF-PBNp-Chi fabrics are found and linked with the functional properties of the fabrics. In this study, the longitudinal and transverse weaves are analyzed with the help of tensile and tear strength for chitosan coated, including the *P. betle* chitosan-coated, fabrics. Prior to washing, the breaking weight of interstices (warp and weft yarn) values for tensile and tear strengths of CF-PBNp-Chi fabric is increased against CF and CF-Chi fabrics. Typically, the comprehensive tensile and tear strength for the fabrics specimen CF-Chi/CF-PBNp-Chi are found to be more than that of CF fabric (Table 2).

This is due to the sharing of the load by the coated nanoparticles on the fabrics leading to an increase in tensile strength. In a similar way, the CF-PBNp-Chi fabric again compliments a greater bursting strength of UC-CF and CF-Chi fabrics. The nanoparticles are strongly followed by the surface of cotton fabrics, leading to an enhancement in crease recovery angle (CRA) of fabrics. The CRA of CF-PBNp-Chi fabric is 104 ± 0.76°, which is more when compared to CF or CF-Chi fabric. The observed greater magnitude of crease recovery is a result of higher absorption of PBNp-Chi nanoparticles on the surface of fabrics without giving any stiffness to the fabric. Apart from this, the CRA of the CF-PBNp-Chi fabrics decreases after 5th and 10th washes due to the reduction of coating with the increase in number of washings.

The blocking for both UV-A (320-400) and UV-B (290–320 nm) is not elucidated for the CF fabrics.[41] As observed in the case of CF-PBNp-Chi fabric, there is a reduction in transmittance of some specific wavelength. This transmittance is the result of blocking UV-B and UV-A radiations. When it comes to coated fabrics, it can be observed that the percentage of UV-B blocking is relatively more when compared to UV-A. The CF-

PBNp-Chi fabric shows great degree of blocking of UV-B radiation when compared to CF-Chi fabrics.[42] Moving ahead, the blocking rate of UV-radiation for CF-PBNp-Chi fabrics after 5th and 10th washes decreases when compared to that of the unwashed cotton fabrics. The reduction in blocking rate of UV-radiation for the coated fabrics elucidated that the subsequent washes entailed the alleviation of a small amount of coating on the surface of fabrics, which is also evidenced from the earlier observations.[43–45]

On the basis of ASTM D6603 standard data, the estimated ultraviolet protection factor (UPF) value for the fabrics that is utilized in UV blocking is much greater than 50. The values for CF and CF-Chi fabrics are lower than 50 when compared to CF-PBNP-Chi fabrics. Table 3 illustrates the UPF value for the CF, CF-Chi, and CF-PBNp-Chi fabric samples. It is quite apparent from Table 3 that CF-PBNp-Chi fabric indicates greater UPF value of about 50, depicting an increased blocking rate of UV-radiation.

The increase in the UV-blocking properties of CF-PBNp-Chi fabric is typically due to the amalgamation of PBNp nanoparticles to colloidal chitosan sol on the surface of cotton fabrics. Thus, the CF-PBNp-Chi fabrics can impeccably combat the UV-radiation after complete washing process.

Antibacterial assessment of coated fabrics

The analysis of the antibacterial activity of the obtained PBNp nanoparticles, including CF, CF-Chi, and CF-PBNp-Chi fabrics, are assessed by cautiously evaluating the diameter of inhibition zone. The disk encumbered with PBNp nanoparticles reflected the maximum zone of inhibition, as compared to *E. coli* and *S. aureus*, at a concentration of 100 mg mL^{-1}. Higher area for the zone of inhibition is noticed for CF-PBNp-Chi fabrics that convenes more inhibitory action (25.45 ± 0.34 and 31.58 ± 0.06 mm) as compared to CF-Chi fabrics. However, when it comes to *S. aureus*, the difference in the magnitude of zone of inhibition is somewhat higher (26.81 ± 0.07 mm and 34.01 ± 0.08 mm) than *E. coli*.
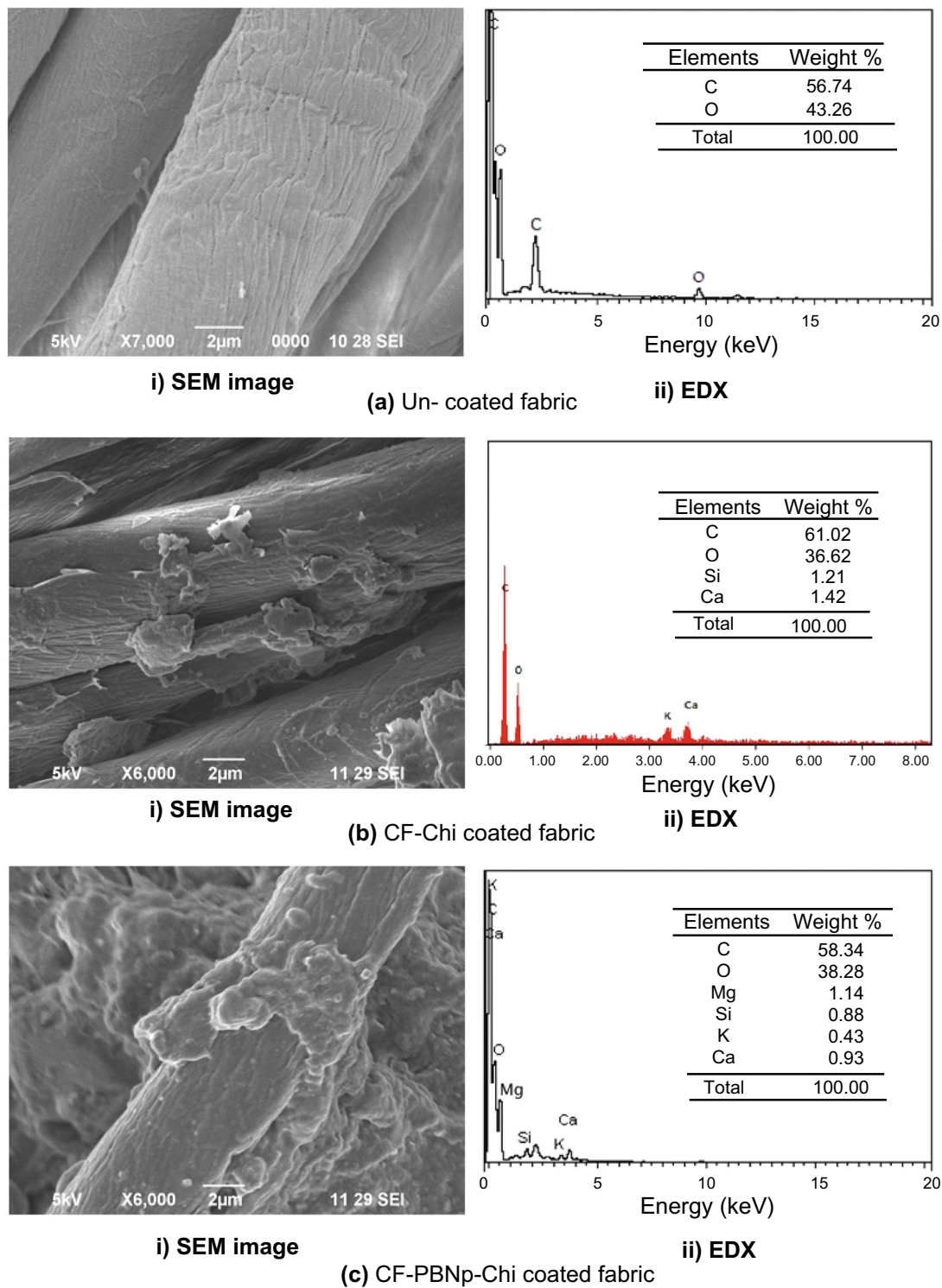
i) SEM image

(a) Un- coated fabric

ii) EDX

Elements	Weight %
C	56.74
O	43.26
Total	100.00

i) SEM image

(b) CF-Chi coated fabric

ii) EDX

Elements	Weight %
C	61.02
O	36.62
Si	1.21
Ca	1.42
Total	100.00

i) SEM image

(c) CF-PBNp-Chi coated fabric

ii) EDX

Elements	Weight %
C	58.34
O	38.28
Mg	1.14
Si	0.88
K	0.43
Ca	0.93
Total	100.00

Fig. 3: SEM images and EDX spectra of (a) uncoated fabric, (b) CF-Chi-coated fabric, and (c) CF-PBNp-Chi-coated fabric before wash

Elements	Weight %
C	61.16
O	36.73
K	1.01
Ca	1.10
Total	100.00

i) SEM image **ii) EDX**

(a) CF-Chi coated fabric (5th time washing)

Elements	Weight %
O	53.13
C	44.99
Mg	0.50
Si	0.72
K	0.23
Ca	0.43
Total	100.00

i) SEM image **ii) EDX**
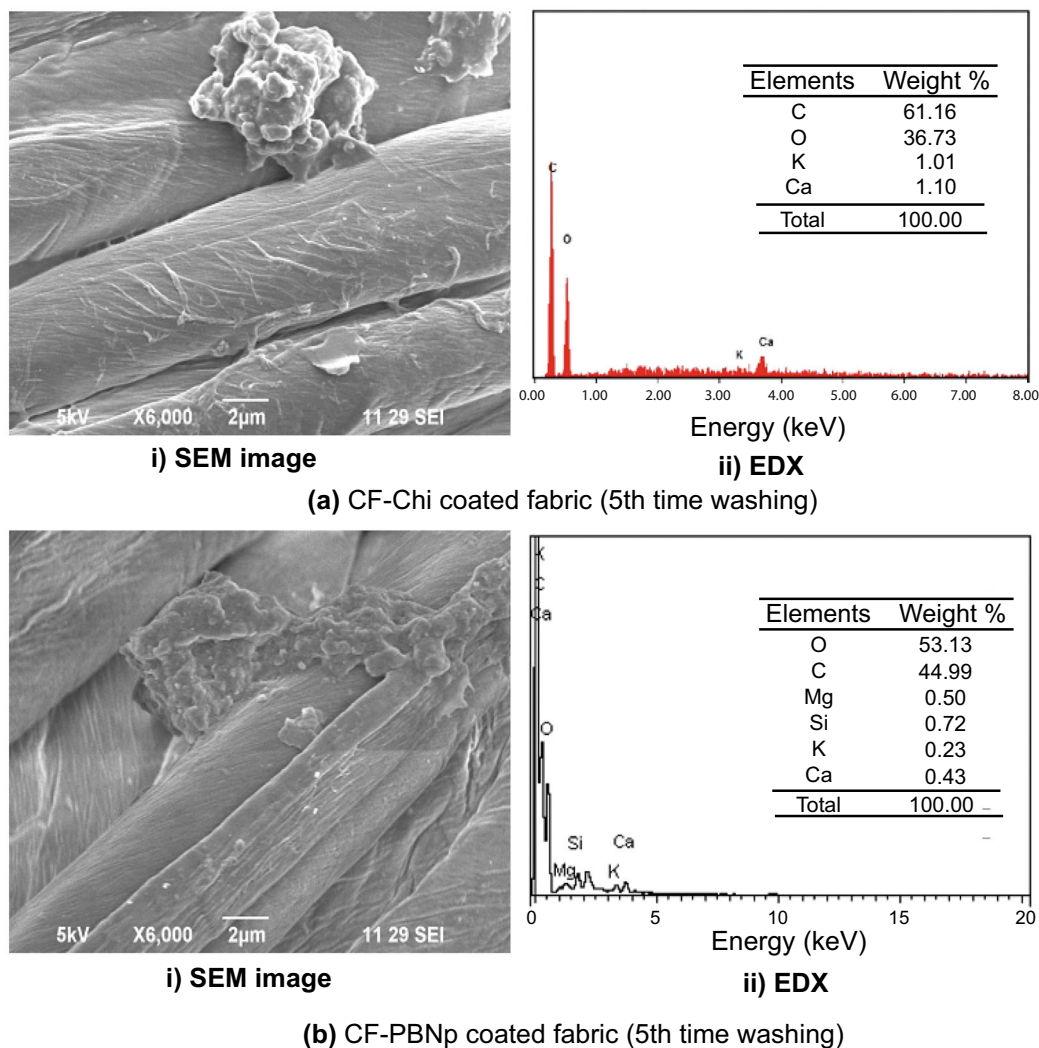
(b) CF-PBNp coated fabric (5th time washing)

Fig. 4: SEM images and EDX spectra of (a) CF-Chi-coated fabric and (b) CF-PBNp-Chi-coated fabric after 5th washing

Additionally, in the case of UC-CF fabrics, the formation of inhibition is completely absent. A thorough analysis of the current outcomes shows that CF-PBNp-Chi fabric displays maximum zone of inhibition against *E. coli* and *S. aureus*. The higher antibacterial action of PBNp sample is obtained due to the existence of phytochemical compounds in *Piper betle* leaves, such as carbohydrates, alkaloids, flavonoids, phenols, protein, and steroids.[46] The above-mentioned bacterial susceptibility investigations coupled by plant extract and herbal nanoparticles show the striking medicinal properties and therapeutic uses of PBNp nanoparticles, which can be utilized in medical textiles.

Percentage reduction test (AATCC 100) and washing durability of coated fabrics

Table 3 shows the quantitative calculation of the antibacterial action for the UC-CF and CF-PBNp-Chi fabrics after percentage reduction test. The percentage reduction test performed against microorganisms treated with CF-PBNp-Chi fabrics both before and after washing is 99% and 93%, respectively. In addition, for CF-Chi fabrics, 77% and 70% are found, respectively. The percentage reduction test does not show any evidence for bactericidal activity of CF fabric. After commencing 5th and 10th wash, the antibacterial activity decreases due to the elimination
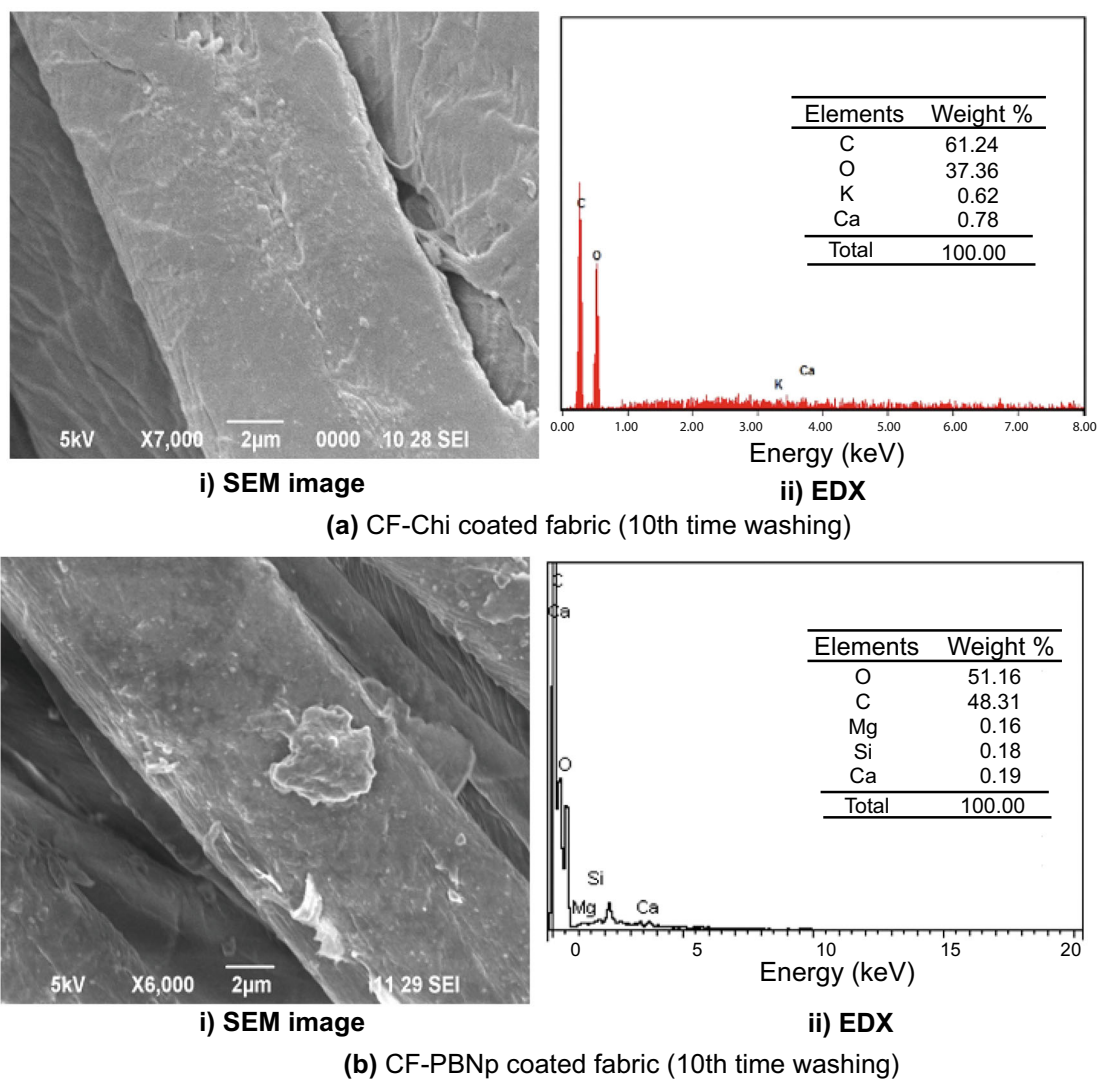
i) SEM image

Elements	Weight %
C	61.24
O	37.36
K	0.62
Ca	0.78
Total	100.00

ii) EDX

(a) CF-Chi coated fabric (10th time washing)

i) SEM image

Elements	Weight %
O	51.16
C	48.31
Mg	0.16
Si	0.18
Ca	0.19
Total	100.00

ii) EDX

(b) CF-PBNp coated fabric (10th time washing)

Fig. 5: SEM images and EDX spectra of (a) CF-Chi-coated fabric and (b) CF-PBNp-Chi-coated fabric after 10th washing

of herbal nanoparticles from fabric surface as compared to *E. coli* and *S. aureus* bacteria. The antibacterial activity of the CF-PBNp-Chi fabrics after 10 washings is more than 40%. The wash durability of CF-PBNp-Chi fabrics after 10th wash is found to be greater, which ensures superior functional properties of herbal nanoparticle-coated fabrics.

Conclusion

Herbal nanoparticles were obtained from the shade-dried leaves of *Piper betle* with the help of ball milling technique that took place in three different milling durations (5, 10, and 15 h). The best sample was collected on the basis of characterization results for further coating on the cotton fabrics. Going ahead, the coated fabrics showed enhanced UV-protective action at 284 nm. A good amount of nanoparticles placed on the fabric surface remained even when it was washed 10 times. The discovered physical properties of the CF-PBNp-Chi-coated fabrics were superior as compared to chitosan-coated or uncoated fabric materials. The herbal nanoparticles on the cotton fabrics also showed better antibacterial activity when compared to *S. aureus* and *E. coli*. From the aforementioned outcomes, it is clear that the natural phytochemical constituents of *P. betle* nanoparticles are good at retaining their medicinal properties and can be coated on cotton fabrics. This, in turn, enhances various properties, like antibacterial activity, washing durability, and UV-blocking ability. Therefore, this study highlights the use of *P. betle* nanoparticles coated with CF-PBNp-Chi fabrics for enhancing applications in biomedical and textile industries.

Table 2: Physical properties of uncoated and nanoparticle chitosan-coated cotton fabrics

Sample code	Tensile strength		Tear strength		Air Permeability (cc s^{-1} cm^{-2})	CRA (W + F)°	Thickness (mm)	Bursting strength (kg cm^{-2})
	Warp (kg)	Weft (kg)	Warp (kg)	Weft (kg)				
UC-CF	46.1 ± 0.1	41.3 ± 0.1	21.2 ± 0.1	17.9 ± 0.1	97.42 ± 0.4	98.6 ± 0.2	0.29 ± 0.1	6.31 ± 0.5
CF-Chi	47.9 ± 0.1	42.8 ± 0.1	23.9 ± 0.2	19.8 ± 0.1	85.97 ± 0.2	103.4 ± 0.4	0.33 ± 0.5	6.62 ± 0.6
CF-PBNp-Chi	48.5 ± 0.4	44.6 ± 0.3	27.1 ± 0.1	22.1 ± 0.1	53.32 ± 0.1	104.8 ± 0.7	0.34 ± 0.1	7.45 ± 0.2
After 5th wash								
UC-CF	33.2 ± 0.1	29.2 ± 0.1	17.3 ± 0.3	13.6 ± 0.2	98.47 ± 0.2	75 ± 0.2	0.27 ± 0.1	6.30 ± 0.1
CF-Chi	35.1 ± 0.1	32.6 ± 0.2	21.4 ± 0.2	14.1 ± 0.1	65.93 ± 0.1	92 ± 0.2	0.32 ± 0.5	6.51 ± 0.1
CF-PBNp-Chi	40.2 ± 0.4	34.3 ± 0.2	23.3 ± 0.1	16.4 ± 0.2	60.02 ± 0.5	96 ± 0.2	0.33 ± 0.1	6.78 ± 0.2
After 10th wash								
UC-CF	26.3 ± 0.3	20.4 ± 0.4	15.4 ± 0.4	10.2 ± 0.4	99.97 ± 0.1	45.6 ± 0.2	0.25 ± 0.1	6.1 ± 0.1
CF-Chi	29.8 ± 0.1	23.1 ± 0.1	16.1 ± 0.1	11.6 ± 0.1	72.44 ± 0.1	61.5 ± 0.1	0.30 ± 0.1	6.3 ± 0.2
CF-PBNp-Chi	29.1 ± 0.1	25.2 ± 0.1	19.5 ± 0.2	13.5 ± 0.2	71.13 ± 0.2	60.8 ± 0.1	0.32 ± 0.2	6.6 ± 0.3

Table 3: Bacterial reduction percentage of *Piper betle* chitosan-coated fabrics

No. of washes	Bacterial reduction percentage (%)				UPF value (%)		
	CF-Chi		CF-PBNp-Chi				
	E. coli	*S. aureus*	*E. coli*	*S. aureus*	UC-CF	CF-Chi	CF-PBNp-Chi
0 wash	82 ± 0.81	78 ± 0.21	95 ± 0.72	99 ± 0.32	13.9	42.8	55.2
5th wash	70 ± 0.31	60 ± 0.11	81 ± 0.34	83 ± 0.39	11.2	40.1	53.8
10th wash	40 ± 0.14	36 ± 0.11	45 ± 0.26	46 ± 0.12	10.8	39.5	50.8

Conflict of interest Authors declare that they have no conflict of interest in this paper.

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