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Original Article

Colorful and antibacterial nylon fabric via *in-situ* biosynthesis of chitosan mediated nanosilver

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ABSTRACT

Herein, functionalization of nylon fabric surfaces in terms of excellent coloration and antibacterial properties were developed by *in-situ* synthesis of chitosan mediated silver nanoparticles (AgNPs). No toxic chemicals were used to ensure eco-friendly conditions. The variation of colors (nearly red, yellow, and blue) was achieved by only regulating precursor concentration and utilization of ascorbic acid. Functionalized nylon fabrics were then characterized by investigating surface morphology, elemental mapping, metal composition, and chemical linkage among compositing components. Results exposed that the particles are within the nano-range size, spherical in shapes, homogenously dispersed over the surfaces, and firmly attached to the nylon fiber by molecular force or double networking properties of chitosan. Color characteristics demonstrate a uniform shade due to the localized surface plasmon resonance (LSPR) properties of AgNPs with brilliant colorfastness and color strength (K/S). The antibacterial properties are found to be significant, with more than 88% bacterial reduction rate against both the gram-positive and gram-negative bacteria even after 20 washing cycles. Overall, this nylon functionalization

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protocol without using traditional chemicals like crosslinkers, binders, or coating agents provides the desired permanent efficiency and safe product.

1. Introduction

The recent research on nanomaterials synthesis and their application for textiles treatment has become a significant interest both from academic and industrial perspectives [1]. The loading of nanomaterials on textile materials means loading new features that can endorse multifunctional properties on them [2,3]. Likewise, the application of silver nanoparticles (AgNPs) on the textile surface can lead to the manufacturing of high-quality products with divergent applications as antibacterial material, self-cleaning, coloration, flame retardancy, hydrophobicity conductive coverage, electronic sensors, photonic, optoelectronic, catalysts and so on [4–7]. In addition, the colorful appearance (which is the primary demand of consumers) of textiles can also be enhanced by the same treatment protocol since novel nanomaterials demonstrate localized surface plasmon resonances (LSPR) [8,9]. In contrast, conventional dyeing processes require massive amounts of chemical ingredients and dyestuffs [10,11]. Generally, traditional dyestuffs contained toxic chromophore (e.g., azo groups), which are severely harmful to the environment and water sources [12–15]. In this regard, biosynthesized nanomaterials-based coloration could be a suitable replacement to manufacture sustainable and functionalized textiles together [16,17]. Synthetic fibers have some significant benefits over natural fibers, i.e., higher strength, better productivity, lower prices, etc. along with some demerits like lower hydrophobicity and dye adhesions [18–22]. Nylon is a synthetic polymer that has significant importance for both the fabric and fibers and after all on textiles as well. Besides, nylon is consumed worldwide for various products such as military uniforms, bags, clothing, and so on. Nanosilver functionalized nylon fabric could be used as a prominent clothing material for army peoples for having protection against bacterial attacks [23].

However, agglomeration is the primary problem for synthesizing metal nanoparticles [24,25]; therefore, a supporting template (i.e., stabilizing and reducing agents) is often required [26–29]. Polysaccharides/carbohydrates [30–32] and phytochemicals of different leaves extract [33,34] are a good example in this case. Chitosan is one of the prominent representatives of polysaccharides, generally obtained from chitin, a homopolymer of β-(1–4) linked N-acetyl-D-glucosamine [35]. This natural polymer can serve as a good template for the synthesis of nanoparticles [36,37]. Consequently, a variety of nanoparticles, including palladium [38], gold [39], silver [36,37], platinum [40], copper [41], etc. were prepared recently. Even, chitosan reduced and stabilized AgNPs were also produced for the coloration and functionalization of textile materials such as cotton [42], linen [43], wool [44], polyester [45], viscose [46], aramid [47], etc. Definitely, chitosan reduced and stabilized AgNPs has not been applied yet for the multi-functionalization of nylon fibers. Therefore, the nano-based treatment of nylon fabric has drawn substantial consideration for improving their typical properties. Although nylon treatment using AgNPs were carried out recently via sonochemical coating [48], ultrasound-assisted coating [49], layer-by-layer deposition [50], surface immobilization [51], and so on but the ultimate protocol and objects were different. Even either they are subjected to complicated processes or toxic chemicals restrict their wider application [52].

Therefore, in this work, a natural reducing and stabilizing agent was used for a one-step *in-situ* synthesis of AgNPs on nylon fabric for colorization and antibacterialization. The formation and deposition of nanosilver on the nylon surfaces were confirmed by scanning electron microscope (SEM), energy-dispersive X-ray (EDX), and elemental mapping. The chemical linkage among compositing components in the resultant product was confirmed by Fourier transfer infrared spectroscopy (FTIR) analysis. Particles were quantitively measured using an X-ray fluorescence (XRF) spectrometer, and cytotoxicity was evaluated by recalculating the amount of AgNPs after standard washes.

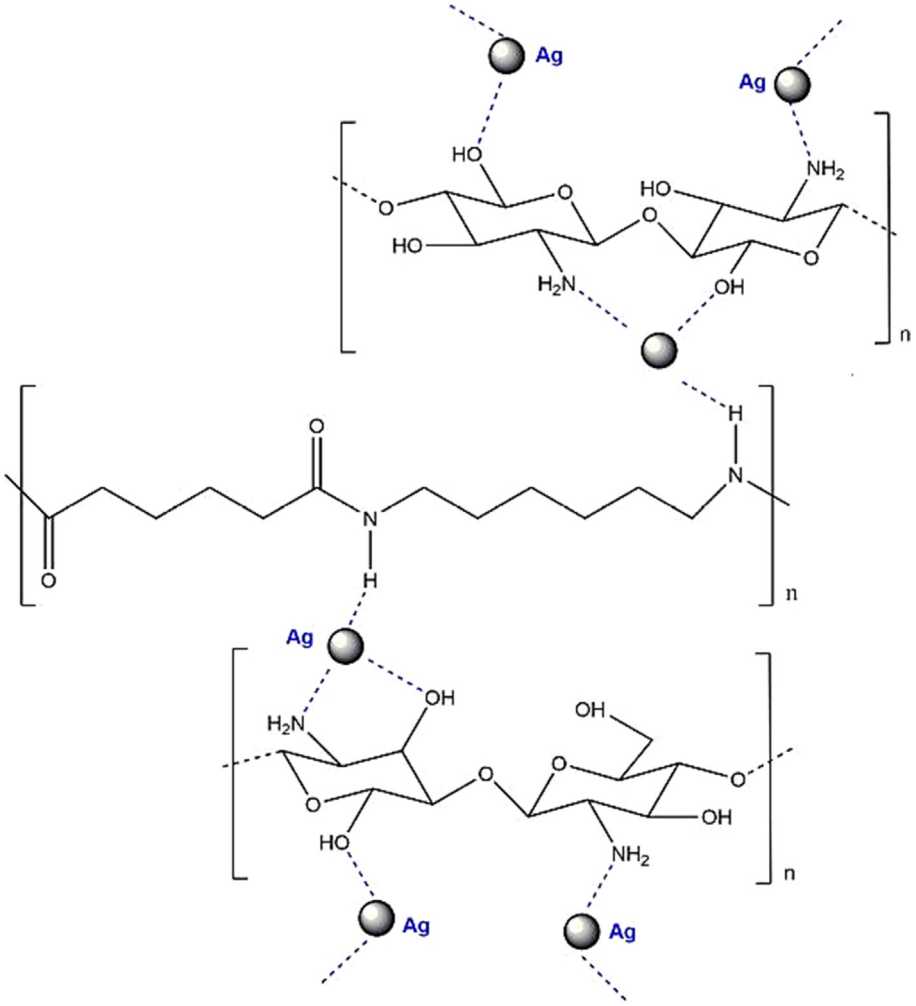
2. Materials and methods

2.1. Materials

Chitosan (molecular weight, Mw $\leq$ 1500 and particle size: 120 mesh), ascorbic acid ($C_6H_6O_8$, 99% purity), and silver nitrate ($AgNO_3$, 99.98% purity) were procured from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China. A plain weave nylon fabric (91 GSM, 70 ends/in., 70 picks/in., and a linear yarn density of 150 denier) was purchased from Suzhou Textile Co., Ltd., Suzhou, China. All of the chemicals and materials were used without any further treatment.

2.2. In-situ *bio-synthesis of AgNPs on nylon*

Prior to the treatment, nylon fabrics were washed through a continuous flow of distilled water at room temperature. During the *in-situ* synthesis of AgNPs on nylon, about 0.65 g of chitosan was primarily mixed in three individual beakers of 100 mL solutions. Approximately 3.4, 1.5, and 3.5 g of $AgNO_3$ were then transferred to beakers for red, yellow, and blue samples, respectively. A certain portion of ascorbic acid (0.65 g) was added to the third beaker for blue-colored nanocolloids. After that, solutions were magnetically stirred at room temperature for 30 min. Then, three pieces of nylon fabrics (10.0 g of each) were individually immersed into the as-prepared solutions by maintaining a materials-to-liquor ratio



Scheme 1 – Possible interaction during the formation and fixation of AgNPs on the nylon fabric.

of 1:20 and stirred for another 30 min to ensure better absorption. Three distinct colors (nearly yellow, red, and blue) on both the solution and fabrics were achieved by raising the temperature to 100 °C for 45 min. All colored samples were then washed and rinsed with tap water for three times to remove any unfixed nano-ingredients from the surface of the materials. Finally, samples were dried in a household tumble dryer at 25 °C temperature for 10 min. All colored fabrics were labeled as R-nylon, Y-nylon, and B-nylon, respectively, for red, yellow, and blue color. The untreated fabrics were also washed and dried in the same protocol to prepare a control sample (C-nylon) for comparison. All samples were stored at standard atmosphere (25 °C temperature and 65% relative humidity) for 24 h before subsequent characterization.

2.3. *Characterization and measurement*

The surface morphologies of both treated and untreated nylon fabrics were carried out by a scanning electron microscope (SEM, S–3400N, Hitachi Ltd., Tokyo, Japan) after the gold coating (E–1045 Ion Sputter, Hitachi Ltd., Tokyo, Japan) at a voltage of 20 kV. The microscope was also equipped with an Oxford Instruments liquid nitrogen (N_2) cooled energy-dispersive X-ray analysis (EDX) detector. An INCA Microanalysis Suite software was used for analyzing the EDX spectra and elemental mapping. The presence of nanoparticles was detected by X-ray fluorescence (XRF, NitonTM XL2, Thermo Scientific™, USA) analyzer. The interaction between the fiber and particle was determined by a Fourier transform infrared (FTIR) spectrophotometer (Bruker Optics, Ettlingen, Germany) within the wavelength range 4000–500 cm^{-1}. The colorimetric values (color difference, color strength and colorfastness) and antibacterial performance (against *Escherichia coli* and *Staphylococcus aureus*) were measured according to our previous report [8].

3. Results and discussions

3.1. In-situ *synthesis of AgNPs on nylon*

Initially, the reaction system was visually observed to detect whether any color changes occur in the nanocolloid. While the system was heated, a colorful appearance of fabrics and solutions was noticed, indicating the formation of AgNPs. The appearance of color specifies the generation of AgNPs on the

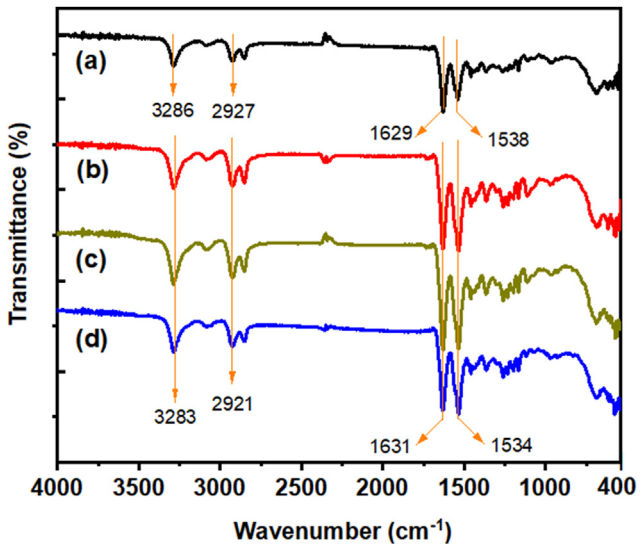


Fig. 1 – FTIR spectra of control and nano-silver treated samples: (a) C-nylon, (b) R-nylon, (c) Y-nylon, and (d) B-nylon.

nylon surface, and this is because of the excitation of surface plasmon vibrations [34]. It has been recognized that the noble metal showed localized surface plasmon resonance (LSPR) by the collective oscillation of conduction electrons in resonance with the wavelength of irradiated light [37]. The diversity of colors was achieved by controlling the precursor's ($AgNO_3$) concentration and applying ascorbic acid since they are major parameters to control the shape and size of synthesized particles. Chitosan, on the other hand, could serve here to reduce silver cation (Ag^+) to neutral ion (Ag^0), as well as to stabilize the produced particles by creating a thin layer of cladding over their surface. However, the particle formation mechanism on the nylon surface can be assumed as (a) when the fabrics were immersed into the ionic solution of silver, they have been absorbed by the nylon surface (nylon–Ag^+) (Eq. (1)). (b) chitosan (CS) aqueous solution was therefore dispersed over the silver-cation absorbed nylon surface to form a ternary complex [nylon–Ag(CS)]$^+$ (Eq. (2)). (c) Finally, this complex further reacted with the OH ions to form nanosilver particles (Eq. (3)). A similar reduction mechanism was also observed for other textile fiber [44]. In addition, there are two different fixation-routes of synthesized particles found for nylon surface treatment. (a) Primarily larger-size particles are entangled inside the fiber/fibril network of the nylon yarn textures. They are then tightly attached to the fiber surface via molecular force developed by the particle's chitosan thin layer. (b) On the other hand, osmotic pressure forces comparatively small particles to penetrate into the fiber porous system (if any) and then entrapped by the chitosan's double network formation capability [47]. However, in both cases, the fixation is achieved through the interaction of Ag^+ ions with amino (-NH_2) and hydroxyl (-OH) groups of nylon and chitosan terminals, respectively (Scheme 1).

$$\text{nylon} + Ag^{*}\,(aq) \rightarrow \text{nylon}{-}Ag^{*} \quad (1)$$

$$\text{nylon}{-}Ag^{*}\,(aq) + \text{CS}\,(aq) \rightarrow [\text{nylon}{-}\text{Ag}\,(\text{CS})]^{*}\,(aq) \quad (2)$$

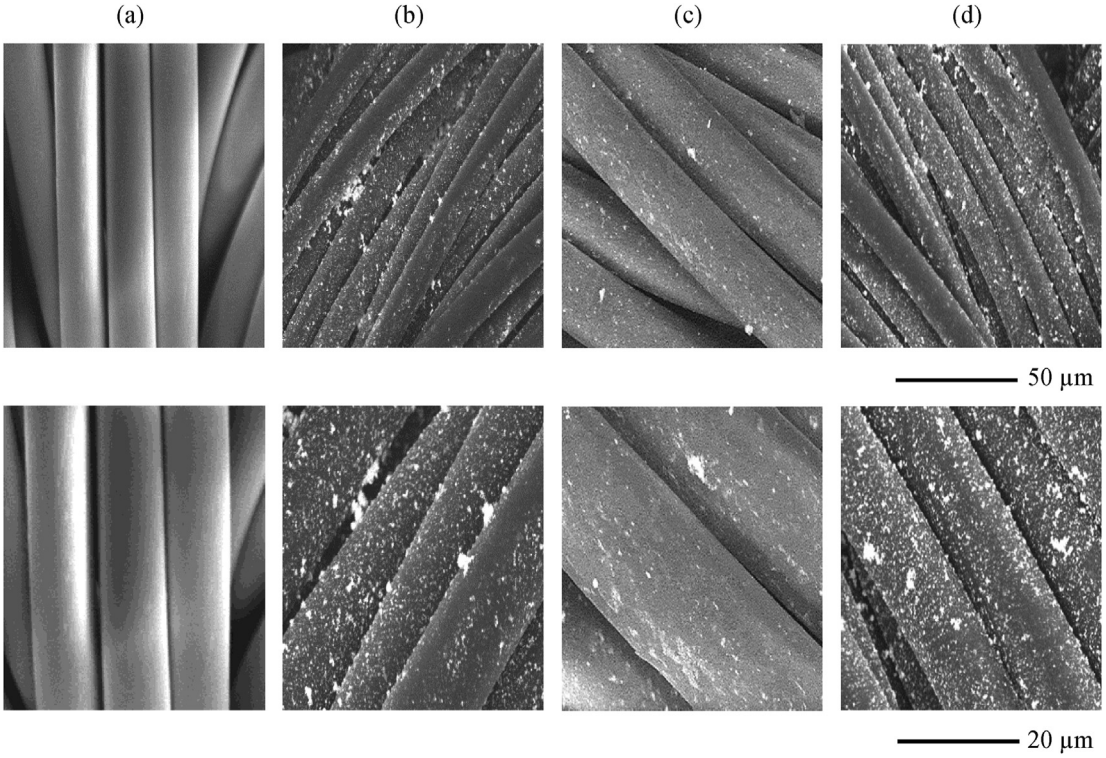


Fig. 2 – Surface morphology of control and nanosilver treated samples: (a) C-nylon, (b) R-nylon, (c) Y-nylon, (d) B-nylon, and their corresponding higher magnification images at the bottom.

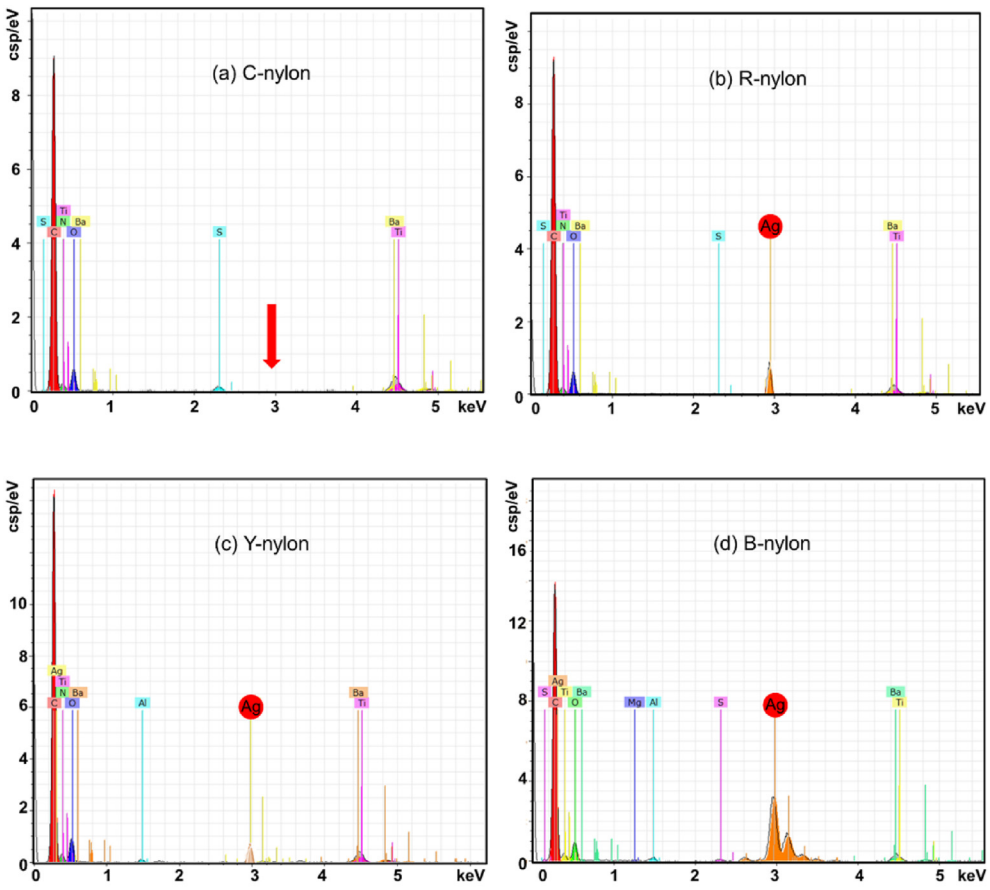


Fig. 3 – EDX liner scanning of control and nanosilver treated samples: (a) C-nylon, (b) R-nylon, (c) Y-nylon, and (d) B-nylon.

$$2\,[\text{nylon}{-}\text{Ag (CS)}]^{+} + 2\,OH^{-} \rightarrow 2\,[\text{nylon}{-}\text{Ag(CS)}]\downarrow + H_2O + 1/2\,O_2 \quad (3)$$

3.2. *Characterization of AgNPs treated nylon*

As a proof concept for the interaction of Ag^{+} ions with nylon and chitosan terminals, Fourier transform infrared spectroscopy (FT-IR) spectra of both treated and control samples were recorded (Fig. 1). The significant peaks of C-nylon (Fig. 1a) display at around 3302 cm^{-1} (amine stretching vibration), 2931 cm^{-1} (CH_2 stretching vibration), 1640 cm^{-1} (amide carbonyl stretching vibration), and 1530 cm^{-1} (N–H bending vibration) wavenumber [53]. This corresponds to the polyamide fiber in the nylon fabric, which is a linear polymer structure formed by linking an amide bond (–CONH–) and many methylenes [53]. However, *in-situ* AgNPs treatment of nylon fabrics causes these peaks dramatically shifted to 3283, 2921, 1631, and 1534 cm^{-1}, respectively (Fig. 1b–d). It can also be observed that the peak intensity for $-NH_2$ and –CONH groups increased significantly for the nanosilver treated fabrics, and this is possibly due to the domination of amine group around chitosan structure. The overall findings indicate that the AgNPs deposition on the fabrics are result of chemical linkages of Ag^{+} to the nylon and chitosan terminals.

The surface morphologies of control and treated samples were observed (Fig. 2). The changes in surface morphology of the nylon caused by the formation and deposition of AgNPs were noticeable. The control nylon showed a smooth and uniform surface (Fig. 2a), whereas the treated fiber demonstrates a rough surface caused by the deposition of AgNPs. It is also observed that the particles are homogeneously distributed over the surface with variable shape (but mostly in spherical) and size, causes the visual color. Comparatively, R-nylon and B-nylon samples showed a significant deposition of particles (Fig. 2b–d). This is due to the use of a higher amount of precursor during treatment, logically forming a higher number of particles. In addition, the treatment neither damages the structure of particles (which in turn shows the bright color) nor the structure of fibers. Overall, the observation ensures that the AgNPs are successfully assembled and visible onto the nylon surface.

Ag elemental signal presence in nano-metallic form rather than Ag compounds was studied by EDX linear scanning (Fig. 3). Evidently, there was no signal detected for Ag in the control samples (Fig. 3a). In contrast, all treated samples showed a distinct presence of the Ag peak at 2.96 keV,

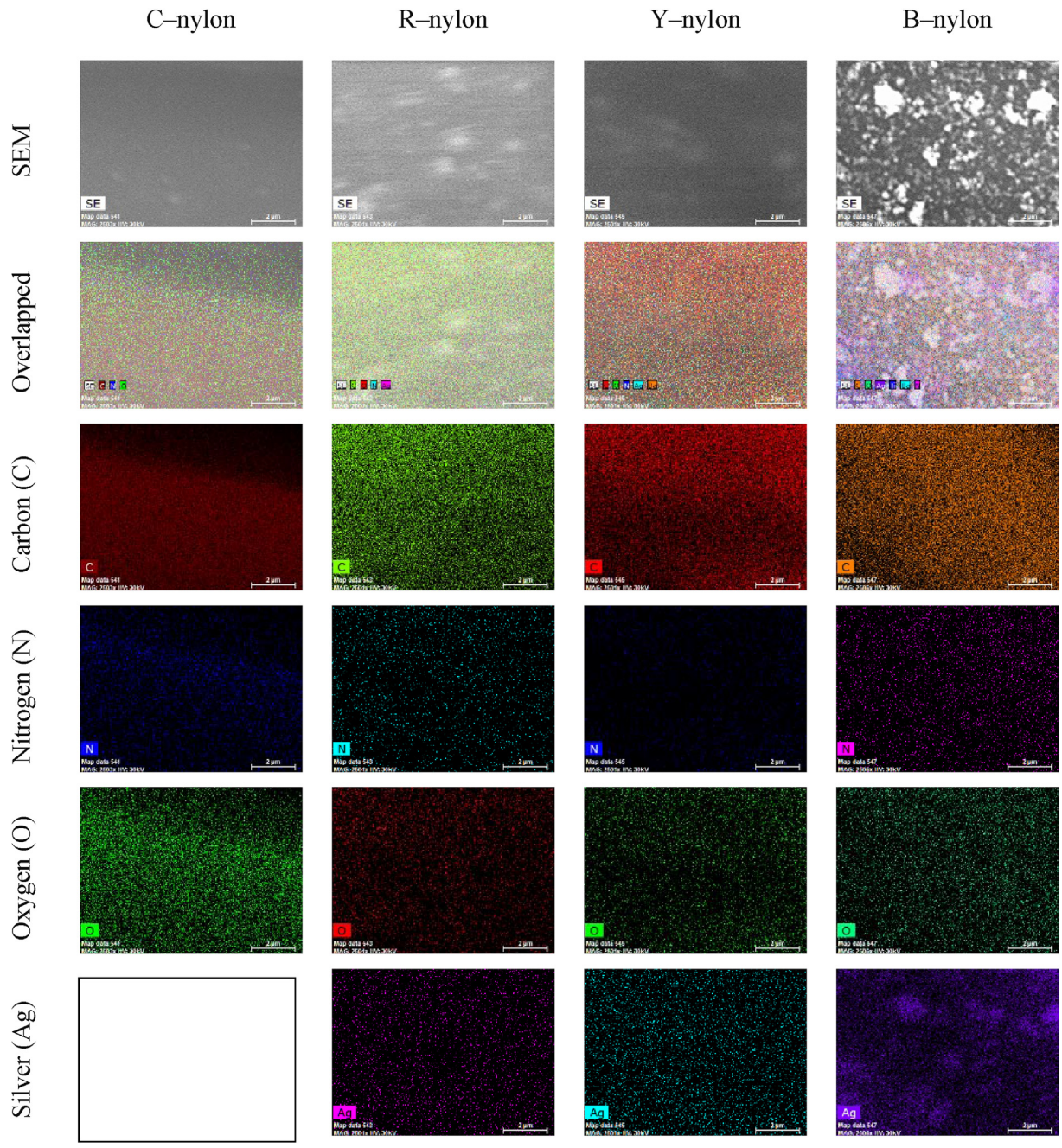


Fig. 4 – SEM-deployed EDX elemental mapping images of control and nanosilver treated samples.

indicating that the Ag has been accurately recognized and exists in the nylon surface (Fig. 3b–d). Hight and exacerbation of the Ag signals were in accordance with the amount of AgNPs content, and the finding is consistent with SEM observation. Moreover, the prominent color difference in the EDX elemental mapping further confirms the existence of AgNPs with homogeneous distribution (Fig. 4). Other bands due to carbon (C), oxygen (O), nitrogen (N) atoms, etc. are also observed both in the EDX line diagram and elemental mapping image. They can be attributed to the chitosan-polysaccharides coating of the AgNPs and nylon molecular structure [33]. In addition, XRF spectrometry was further used to analyses the elemental existence before and after the treatment (Table 1a). The results revealed that about 445, 388, and 474 ppm particles are deposited on the R-nylon, Y-nylon, and B-nylon, respectively. Overall, it could be concluded that the *in-situ* synthesis of AgNPs caused a significant amount of particle deposition on the surface of the fibers.

The term "human safety" comes in the front line while talking about silver due to its traditional toxicity. Hence, it is significant to measure the quantity of Ag content in the treated fabrics and the amount of Ag released after the standard washing procedure. Therefore, the Ag content in the treated fabrics was calculated after 20 washing cycles using the same XRF instrument. It was found that approximately 251, 202, and 285 ppm Ag remained in the R-nylon, Y-nylon,

Table 1 – XRF measurements, color difference, fastness and color strength values.

Samples	(a) XRF measurements		ΔE	(b) Color difference, fastness and color strength				
	$AgNO_3$/CS/AA	Ag content (ppm)		K/S	CFW	CFL	CFR (wet)	CFR (dry)
R-nylon	3.4/0.6/-	445 ± 11	26.5	1.266	4–5	5	3	3–4
Y-nylon	1.5/0.6/-	388 ± 10	18.4	1.321	4	5–6	3	4–5
B-nylon	3.5/0.6/0.65	474 ± 18	29.9	1.293	4	5	2–3	4

CS – chitosan; AA – ascorbic acid; ΔE – color difference, K/S – color strength; CFW – colorfastness to wash; CFL – colorfastness to light; and CFR – colorfastness to rubbing.

and B-nylon, respectively. It indicates, approximately 195 ppm Ag goes away by 20 cycles of washing, which is calculated to be 9.75 ppm per wash. However, the AgNPs are released to the liquid media in a concentration lower than 10 ppm, far below the toxicity limit for humans [54]. In addition, as-prepared particles are well coated by a thin layer of nontoxic chitosan cladding. Accordingly, it can be recommended that the current approach for nylon treatment via *in-situ* synthesis of AgNPs is safe in terms of human toxicity.

3.3. *Color strength and fastness*

The LSPR properties of AgNPs led to brilliant and fascinating multiple colored nylon fabrics through *in-situ* treatment without introducing traditional dyes [45]. The photographs of the nylon fabrics before and after chitosan assisted *in-situ* synthesis of AgNPs are shown in Fig. 5. It can be observed that the white/grey nylon fabrics changed to nearly red, yellow, or blue color (depending on the concentration of precursor and use of ascorbic acid) due to the AgNPs deposition on the nylon surface. Therefore, the color difference (ΔE) was quantitatively measured using the CIE L*a*b* coordinates and found to be 26.5, 18.4, and 29.9 for R-nylon, Y-nylon, and B-nylon, respectively (Table 1b). Similarly, color strength (K/S) values were also measured and observed that the K/S value increased from theoretical zero value (for C-nylon) to 1.266, 1.321, and 1.293 for R-nylon, Y-nylon, and B-nylon, respectively. Generally, a textile product has gone through essential storage from the production house to consumer end-use. Therefore, the storage ability of the treated fabric was evaluated by re-testing the K/S value after one year of storage at standard atmosphere. The K/S values were found to 1.201, 1.222, and 1.218 for R-nylon, Y-nylon, and B-nylon, respectively, which are very close to the values obtained for freshly prepared samples. It indicates that the atmospheric environment does not influence the color properties and bonding of AgNPs to the fabric surface in terms of one-year storage.

The term 'coloration' is not limited by merely imparting color on the substance, and obviously, they should be withstanding under certain conditions. Therefore, the color withstanding of the treated fabrics were assessed in terms of colorfastness to light (CFL), wash (CFW), and rubbing (CFR). The results are numerically expressed in Table 1 and shown good to excellent fastness ratings for all tread samples. For example, the CFL rating within the range of 5–6 indicating 'excellent' color stability against light. It means the treated fabrics cannot take part in any major photo-degradation upon exposure to sunlight. Similarly, CFW ratings equal to or higher than 4, indicating 'good' color stability in the laundry wash. Moreover, CFR rating (both in wet and dry conditions) ranging from 2 to 5, meaning 'fair' to 'excellent' color stability under rubbing condition. According to the Chinese National Standards for Textiles, the overall results are acceptable and meet commercial requirements [45]. It is worth noting, as-studied fastness ratings ranged from good to excellent for all the treated samples are attributed to chemically stable and durable interactions between chitosan-coated AgNPs and surface functional groups of nylon. Therefore, the current coloration process via *in-situ* synthesis of AgNPs offers a simple but effective nylon-dyeing technique without using traditional dyes and auxiliaries like binders, crosslinkers, or coating agent. While acquiring the desired efficiency in the conventional dyeing and finishing of synthetic fibers, AgNPs-functionalization imparts the brilliant colors and withstand the color under primary wearable conditions.

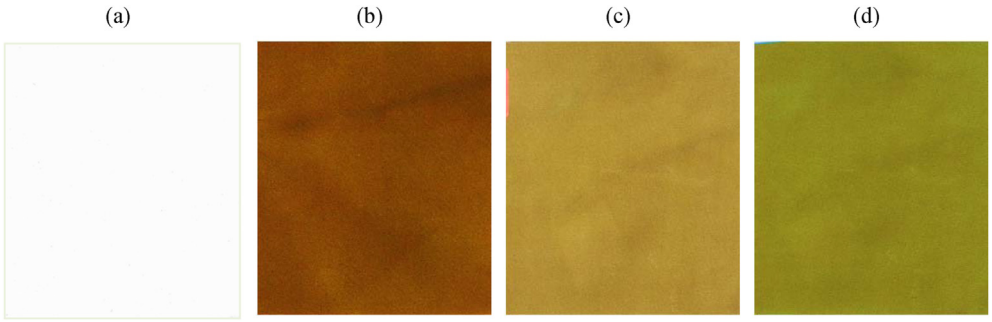


Fig. 5 – Visual appearance of control and nanosilver treated samples: (a) C-nylon, (b) R-nylon, (c) Y-nylon, and (d) B-nylon.

3.4. *Antibacterial functions*

The antibacterial action of AgNPs modified nylon fabrics was evaluated by the disk diffusion test method and found to be a substantial indication of antibacterial effects. The inhabitation zone (mm) created around the tested fabric on the agar-plate was measured for antibacterial analysis (Fig. 6a_1–a_2). Here, *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*) were used as gram-positive and gram-negative bacteria. It is quite clear, the *in-situ* deposition of AgNPs into nylon fabrics have remarkably enhanced antibacterial functions against both pathogen bacteria and created around 25 mm transparent inhabitation zone for all samples. The quantitative data in terms of bacterial reduction (%) are also calculated (Fig. 6b_1–b_2) and found more than 97% reduction for all the treated nylon fabrics. In addition, enhancement of antibacterial function in the fabrics should have sufficient laundering durability since they are subjected to repeated wash in the practical use as textile. Therefore, the AgNPs deposited nylon fabrics were washed through a certain number of laundering cycles, and their antibacterial performances were studied. Both zones of inhabitation and bacterial reduction rates of the treated nylon samples were recorded after 5, 10, 15, and 20 washing cycles. It was observed that the treated nylon to each 5-washing cycle leads to values slightly decreased in both the inhabitation zone and bacterial reduction rate. Interestingly,

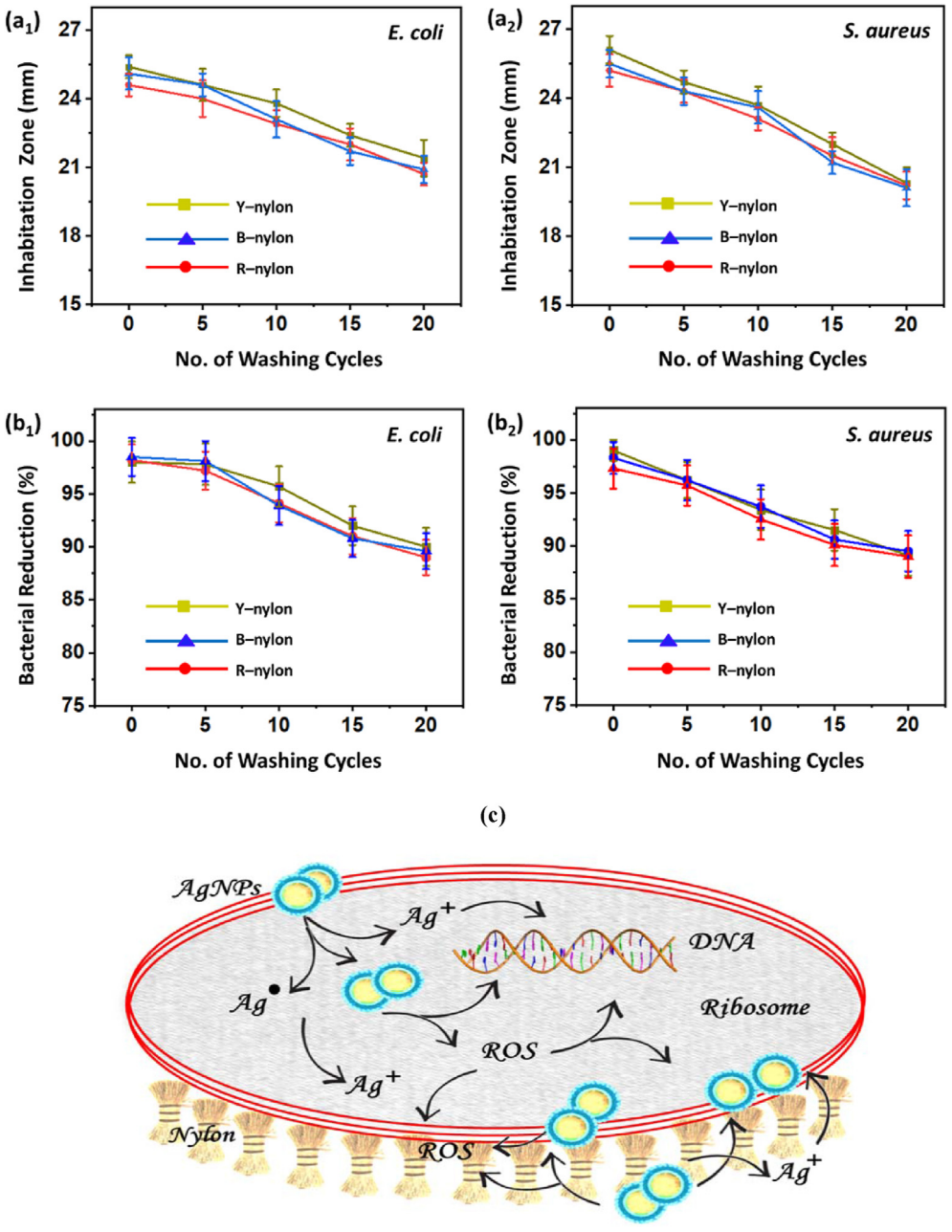


Fig. 6 – Antibacterial performance of nanosilver treated fabrics: zone of inhabitation (a_1, a_2), and rate of bacterial reduction (b_1, b_2) for *E. coli* (a_1, b_1) and *S. aureus* (a_2, b_2). (c) Schematic representation for antibacterial mechanism.

the inhabitation zone was remaining over 20 mm with a bacterial reduction rate of more than 88% for both *E. coli* and *S. aureus* even after 20 washing cycles.

The antibacterial action of silver ions on microorganisms is not unknown yet [55]. Two possible mechanisms can be worked behind the antibacterial activity of AgNPs treated nylon fabric. (1) During the formation of AgNPs, the oxidation of neutral silver ion (Ag^0) to cation (Ag^+) happened to a great extent because of their high sensitivity to oxygen or water molecules. Accordingly, the extreme affinity of those cations to sulfur (S) or phosphorous (P) is considered to be a primary way of antibacterial effect. Mostly, both inner and outer sides of bacterial cell membranes consist of abundant S-containing protein, where AgNPs/Ag + can efficiently react to affect the bacterial cell functionality. Later on, they can react with P-moieties in DNA, resulting in the stopping of DNA replication, leading to the hindering of enzyme functions [56]. (2) The Ag + derived by AgNPs can also catalyze the generation of oxygen radicals ($H_2O + 1/2\ O_2 \rightarrow H_2O_2 \rightarrow H_2O + O\bullet$) that oxidize the bacteria's molecular structure. It is worth noting; such a mechanism does not require any direct interaction between the antibacterial agent (Ag^+) and pathogen because the generated reactive oxygen species (ROS) disperses from the fabric to the neighboring atmosphere. Silver ions (Ag^+) can lead to bacterial cell death due to the denaturation of protein through their interaction with the nucleophilic amino acid of proteins and attach to sulfhydryl, amino, imidazole, phosphate and carboxyl groups of membrane or enzyme proteins [56]. Accordingly, a schematic illustration for the antibacterial mechanism of the AgNPs treated nylon is shown in Fig. 6c.

However, the growth of AgNPs onto nylon fabrics provides excellent colored properties as well as an antibacterial function against both gram-positive and gram-negative bacteria. The excellent laundering durability in terms of colorfastness to wash and bacterial reduction (%) is attributed to the strong bonding between AgNPs and nylon surfaces through *in-situ* synthesis. These collective outcomes confirmed that the straightforward deposition of AgNPs is effectively applicable for colored nylon fabrics production with bacterial protective properties. The conventional chemical reduction technique is associated with environmental and human toxicity. Our toxic reagentless approach is eco-friendly, safer in terms of human toxicity while used as wearable cloth, and industrially feasible.

4. Conclusions

A facile and straightforward technique is applied here to produce colorful and antibacterial nylon fabrics via *in-situ* synthesis of silver nanoparticles. The surface plasmon property of nanosilver imparted nylon with multiple colors, which can be controlled by adjusting the precursor content and ascorbic acid on nylon. The surface characterization showed that the AgNPs are mostly spherical, and their homogeneous distribution influenced the ultimate morphology of nylon fibers. The successful deposition of particles was achieved through the molecular force developed by chitosan's thin layer over the particles and its double network formation capability via chemical linkage. Quantitative measurements using XRF for the presence/release of AgNPs by each cycle of washes and their comparison with available standards revealed that the approach is safe in terms of human toxicity. Due to the traditional features of metallic silver, treated nylon demonstrate a high level of antibacterial properties even after 20-cycles of standard washing (up to 88% reduction) against both gram-positive and negative strains. The colored nylon fabrics caused by nanosilver deposition possessed good colorfastness (to light, wash, and rubbing) and color strength. It can facilitate the practical implementation of *in-situ* coloration involving noble metal nanoparticles and overcome the limitations of traditional dyeing technology.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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