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*Research Article*

# *In Situ* Growth of ZnO Nanostructures on Cotton Fabric by Solochemical Process for Antibacterial Purposes

**D. A. R. Souza,[1] M. Gusatti,[1] R. Z. Ternus,[2] M. A. Fiori,[2] and H. G. Riella[1]**

[1]*Laboratório de Materiais e Corrosão, Departamento de Engenharia Química e Engenharia de Alimentos, Programa de Pós-Graduação em Engenharia Química, Universidade Federal de Santa Catarina, Campus Universitário, Florianópolis, Santa Catarina, P.O. Box: 476, 88040-900, Brazil*
[2]*Departamento de Engenharia Química, Programa de Pós-Graduação em Ciências Ambientais e de Tecnologia e Gestão da Inovação, Universidade Comunitária da Região de Chapecó, Chapecó, Santa Catarina, P. O. Box: 1141, 89809-000, Brazil*

Correspondence should be addressed to M. Gusatti; marivoneg@gmail.com

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A practical and economical method was developed for the production of an antibacterial cotton fabric using zinc oxide nanostructures without the use of surface modifying agents. In this process, zinc nitrate hexahydrate and potassium hydroxide were used as starting materials and the reaction was performed at 50°C. The in situ growth of ZnO nanostructures on cotton fabric occurred in a single-stage process, and it started when the fabric samples were dipped for 1 min in the solution containing all the starting materials. The treated and untreated fabric samples were characterized by X-ray diffraction (XRD), transmission electron microscopy (TEM), scanning electron microscopy (SEM), and EDS. The cotton fabrics coated with ZnO nanostructures presented an antibacterial efficiency towards *Pseudomonas aeruginosa*, a gram-negative bacteria, and *Staphylococcus aureus* (*S. aureus*), a gram-positive bacteria.

## 1. Introduction

In recent years, much effort has been devoted to the application of metal oxide nanoparticles to textile materials for the development of functional fabrics with novel and improved properties [1–5]. These new fabrics derived from natural raw materials have potential uses in several technological areas and a high added value in comparison to conventional textiles [6, 7].

Among the metal oxides used for the preparation of treated fabrics, ZnO nanostructures have a great application potential due to their unique properties such as chemical stability, nontoxicity, antibacterial and UV-protection, high photocatalytic activity, and high transparency in the visible wavelength range [8–13]. Moreover, this material possesses some advantages over silver nanoparticles, such as UV-blocking property and lower cost, and it does not cause a change in the color of the final product [14–16]. Owing to these remarkable properties, ZnO nanoparticles can be directly used for medical and textile applications such as in cosmetics, elimination of pollution, and protective medical clothes [17–23].

ZnO nanostructures have been applied on textile materials through different methods such as hydrothermal route, layer-by-layer deposition, pad-dry procedure, ultrasonic irradiation technique, and sol-gel process [24–28]. Some of these methods are expensive and carried out by using high temperatures, advanced technical equipment, complex procedure, and several steps [4]. In this study, we describe an in situ growth of ZnO nanostructures on cotton fabric through a simple solochemical process. Up to now, the solochemical technique has been standing out in the preparation of ZnO nanocrystals with high purity, especially at low temperatures [29–32]. This method is based on chemical reactions involving an alkaline material, such as $NH_4OH$ [29, 30] and NaOH [31, 32], and a zinc precursor under controlled temperature and a slow mixture of the reagents. In this process, the ZnO nanocrystals are produced without

pretreatment of the reactants or calcination. In addition, the reactions take place without any additional reagent or additive. Thus, this technique is an efficient and very simple approach to the production of ZnO nanocrystals.

Here, the solochemical process was used for the first time for the in situ growth of ZnO nanoparticles on the cotton substrate to obtain an antibacterial textile material. This processing basically consisted of dipping the cotton fabric in a reactional medium at 50°C during a short period of 1 min. The dipping of the textile substrates in the solution was performed at different aging times of the reaction (0 h, 1 h, and 2 h). Thus, it was possible to evaluate the influence of the aging time on the ZnO structural properties and antibacterial activity of the fabrics treated in the solochemical process. Therefore, in this study, we are going to demonstrate that an antibacterial cotton fabric can be prepared by a rapid and economical solochemical method without any further process. Thus, this route can be a viable substitute for other conventional approaches to the in situ growth of ZnO nanostructures on the cotton fibers.

## 2. Experimental Procedure

*2.1. Presynthesis.* The cotton fabrics with 148 g/m$^2$ used in this research were supplied by a local store. This cotton fabric was washed with hot distilled water at 100°C for 20 min to remove any surface contaminants. Then, the cotton fabric was rinsed with distilled water and dried at room temperature.

*2.2. In Situ Synthesis of ZnO Nanostructures on Cotton Fabrics.* The solochemical synthesis was performed with an experimental procedure similar to that reported in [31]. However, in this study, the ZnO nanocrystals were prepared using zinc nitrate hexahydrate ($Zn(NO_3)_2{\cdot}6H_2O$) and potassium hydroxide (KOH) as raw materials. These reagents were used without further purification and dissolved in deionized water.

The procedure started when a 1.0 M alkaline solution was fed into a reactor and stirred until it reached the desired temperature (50°C). At this time, a 0.7 M zinc precursor solution, at room temperature, was added into the reactor by dripping for 30 min, maintaining the same temperature and agitation conditions during the entire experimental procedure.

At the end of dripping (0 h), a dried cotton fabric sample (A) was dipped into the reactional solution where it remained for a short period of 1 min. Thereafter, this fabric sample was removed from the solution and the reaction proceeded under the same temperature and stirring conditions for 1 h. At this time, a new fabric sample (B) was dipped into the reactional solution for 1 min. Thus, sample B was removed from the solution and the reaction proceeded for more 1 h at 50°C. When the reaction reaches an aging time of 2 h, the last cotton sample (C) was dipped into the reactional solution, where it also remained for only 1 min. After being removed from the reactor, all cotton fabric samples were dried in a vacuum oven at 150°C for 10 min.

The characterization of the fabric samples was done by X-ray diffraction (XRD) with a Philips X'Pert diffractometer using Cu K$\alpha$ ($\lambda = 1.54$ Å) as incident radiation, operating at 40 kV and 30 mA. The surface morphology of the cotton fabric samples was examined by scanning electron microscopy (SEM, JEOL JSM-6701) equipped with energy dispersive X-ray spectroscopy (EDS). Transmission electron microscopy was carried out on a JEM-1011 microscope with the accelerating voltage of 100 kV. For the TEM study, the powder formed on the cotton samples was extracted from the fabric surface using ultrasonication and isopropyl alcohol (IPA). A drop of each solution was taken on a carbon grid for TEM imaging, which was purchased commercially.

*2.3. Antibacterial Tests.* Antibacterial activity of treated and untreated samples was determined against *Staphylococcus aureus* (ATCC 25923) and *Pseudomonas aeruginosa* (ATCC 27853). The evaluation of antimicrobial activity by diffusion in solid medium was based on a qualitative method NCCLS (2003). In this process, fresh bacterial strains were inoculated on nutrient agar (24 h at 35°C). Bacterial suspension was prepared in a saline solution (0.9%) at a concentration of about $10^8$ UFC·mL$^{-1}$.

Subsequently, a dilution of the bacterial suspension was prepared and it was equal to $10^4$ UFC·mL$^{-1}$. The treated and untreated cotton fabric samples with a diameter of 7 mm were placed on the seeded agar surface. The plates were incubated in a bacteriological oven (18 h at 35°C). Then, the halos were measured using a metric ruler. The qualitative evaluation is performed in the first zone (halo) of growth inhibition. The halo is measured starting from the edge of the fabric sample to the border of the beginning of the growth of microorganisms.

Antibacterial activity of treated and untreated samples was also determined following the Japanese Industrial Standard (JIS Z 2801: 2010). Fabrics were separately inoculated with 40 $\mu$L of a bacterial suspension containing $10^4$ UFC·mL$^{-1}$. Plates were incubated for 0 h, 2 h, 5 h, 10 h, and 24 h at 35°C. Then, the samples were diluted with 10 mL of 1/500 Luria Bertani (LB) broth and stirred for 120 s. Decimal dilutions were performed for determining the number of viable bacterial cells. 1.0 mL of each of these dilutions was inoculated into sterile petri plates and homogenized with 15 mL of agar for bacterial counts. After solidification, the petri plates were inverted and incubated at $35 \pm 1$°C for 48 h. Subsequently, the number of colonies was counted and the result was expressed in a number of viable bacteria.

## 3. Results and Discussion

*3.1. XRD Results.* Figure 1 shows the X-ray diffraction patterns of the cotton fabric samples (A, B, and C) that were dipped for 1 min into a $Zn(NO_3)_2{\cdot}6H_2O$ and KOH mixed solution at different aging times (0 h, 1 h, and 2 h).

As can be seen, there is no significant difference between XRD patterns of the fabric samples treated at different reaction times. The diffraction peaks at $2\theta$ values of 14.78°, 16.49°, and 22.76° corresponding to the diffractions planes (1 0 1), (1 0 $\bar{1}$), and (0 0 2) are present in the structure of the cellulosic cotton fiber [33]. The other diffraction peaks ($2\theta = 31.84°$, 34.54°, 36.29°, 47.58°, 56.71°, 62.98°, and

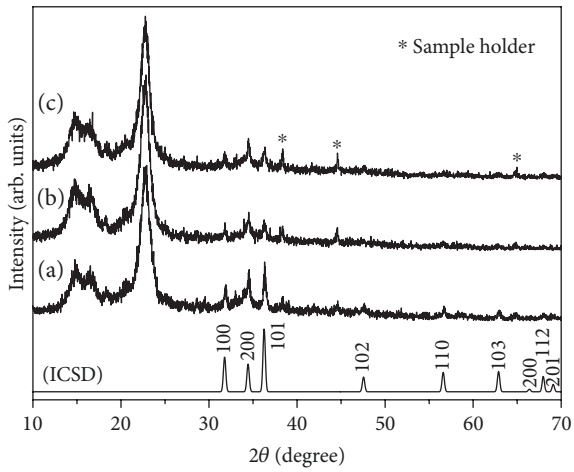


FIGURE 1: XRD patterns of the treated fabric samples after (a) 0 h, (b) 1 h, and (c) 2 h of reaction. Dummy pattern of the ICSD card no. 57450 for the hexagonal ZnO with a wurtzite structure is also presented.

67.98°) present in the XRD patterns can be indexed to the hexagonal ZnO with a wurtzite structure (ICSD card No. 57450). These results indicated the successful formation of ZnO on the surface of the cotton fabric samples treated in the solochemical process.

The crystallite size of the ZnO present on the cotton fabrics was calculated from its higher intense peak in the XRD patterns using Debye Scherrer's equation (i.e., $D = (K * \lambda)/(\beta * \cos\theta)$, where $D$ is the average crystallite size, $\lambda$ is the X-ray wavelength of Cu K$\alpha$ radiation ($\lambda = 1.54$ Å), $K$ is a constant taken as 0.89, $\theta$ is the diffraction angle, and $\beta$ is the full width at half-maximum (FWHM) in radians [34]). For sample A, the peak used to calculate the crystallite size was (1 0 1). For samples B and C, the crystallite sizes were obtained using the diffraction peak (0 0 2). The estimated average crystallite sizes were 28.83 nm, 30.68 nm, and 24.09 nm for the ZnO structures formed on the surface of the cotton fabrics after 0 h, 1 h, and 2 h of solochemical reaction, respectively.

*3.2. SEM and EDS Results.* Figure 2 shows the SEM images of the treated and untreated cotton fabric samples. As can be seen in these images, there is a clear difference between the treated and untreated cotton substrates in terms of the morphology of the fiber surfaces. The untreated cotton fibers exhibit a smooth texture with some cavities (Figure 2(a)). On the other hand, the fabrics treated in the reaction solution at different aging times have several ZnO structures on the surface of their fibers (Figures 2(b)–2(d)).

As we can see in Figure 2(b)-($b_1$), the ZnO structures formed on the surface of the cotton sample dipped in the reactional medium at the beginning of the reaction (0 h) present a plate-like morphology with a high degree of particle agglomeration.

On the other hand, SEM images of the ZnO structures formed on the surface of the fabric dipped in the solution at the aging time of 1 h (Figure 2(c)-($c_1$)) show the presence of some large ZnO plates on the surface of the cotton fibers. On these large ZnO plates, agglomerates composed of several smaller ZnO nanostructures can also be visualized.

Finally, the immersion of the cotton fabric in the solution after 2 h of reaction (Figure 2(d)-($d_1$)) led to the formation of ZnO nanostructures with a size and shape apparently more homogeneous and better dispersed on the surface of the cotton fibers than the previous samples.

The EDS spectra of the untreated and treated cotton fabric samples are shown in Figures 3(a)–3(d). The EDS spectrum of the untreated fabric showed no presence of the zinc element or other new elements (Figure 3(a)). On the other hand, the EDS results confirmed the presence of Zn and O elements related to ZnO nanostructures on the cotton samples treated during immersion of the fabrics in the solochemical solution at different aging times (Figures 3(b)–3(d)).

*3.3. TEM Results.* The TEM micrographs and the selected area electron diffraction (SAED) patterns of the ZnO nanostructures formed on the cotton fabrics are shown in Figure 4. This TEM analysis was performed with the ZnO powder extracted from the cotton fabric samples. TEM images (Figure 4) show that the ZnO nanostructures formed on the surface of the fibers at different aging times have different morphologies, and the predominant shapes are rods, plates, and rounded agglomerates with a considerable degree of agglomeration.

The TEM image of the ZnO nanostructures grown on the fabric that was immersed in the solution with an aging time of 0 h shows the presence of short nanorods and rounded nanostructures with an average diameter of about $16 \pm 3$ nm (Figure 4(a)-($a_1$)).

On the other hand, the immersion of the cotton substrate in the reactional solution after 1 h of reaction resulted in ZnO clusters with larger sizes and rod-like and plate-like morphologies (Figure 4(b)-($b_1$)). The ZnO nanostructures formed under this condition have an average diameter of about $28 \pm 7$ nm.

Finally, the ZnO nanostructures grown in the fabric dipped in the solochemical solution with an aging time of 2 h (Figure 4(c)-($c_1$)) have an average diameter that is slightly smaller ($23 \pm 6$ nm) than those formed at an aging time of 1 h. In general, rod-like and plate-like structures are the predominant morphology of the ZnO structures produced at a longer aging time of the reaction.

The selected area electron diffraction (SAED) patterns (inset in Figure 4 ($a_1$–$c_1$)) suggest that the ZnO nanostructures obtained on the surface of the cotton fibers during the solochemical processing at all aging times of the reaction are crystalline in nature.

*3.4. Possible In Situ Growth Mechanism.* According to our results, the dipping of the cotton samples in the solochemical reactional medium at 50°C for a short period of 1 min was sufficient to promote the *in situ* growth of ZnO nanostructures on the cotton fibers at all aging times studied. SEM micrographs of the samples showed that in shorter aging times (0 h and 1 h), few but large ZnO structures were present

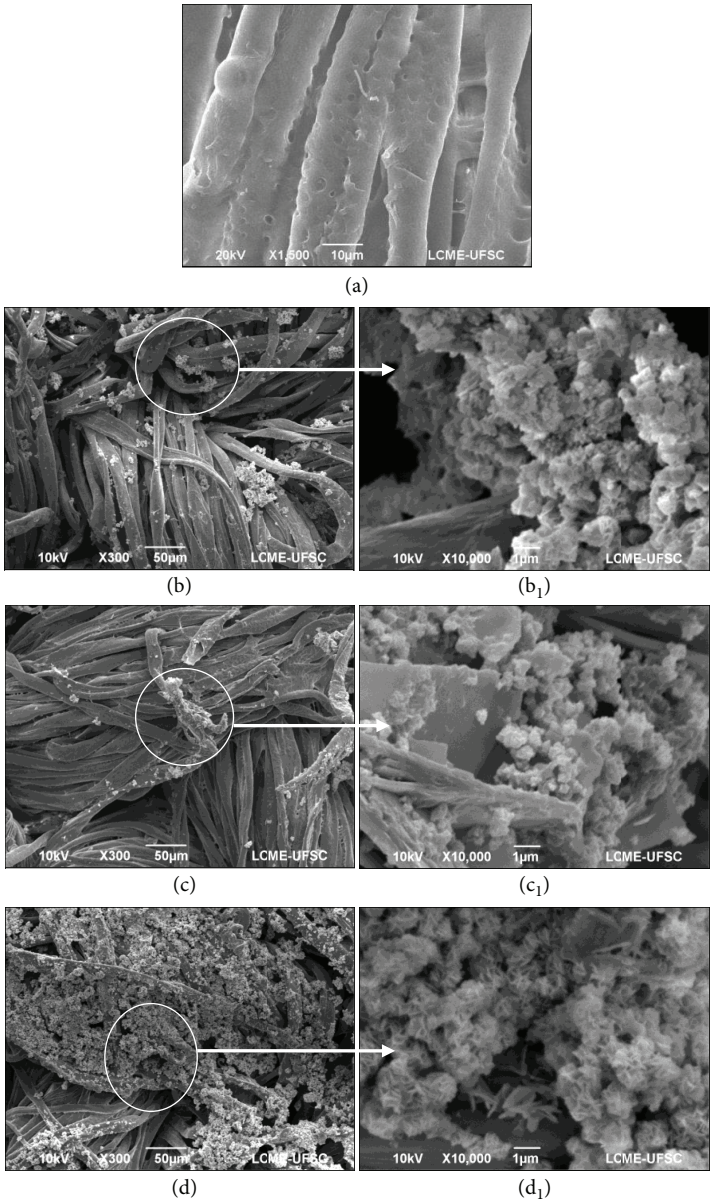


Figure 2: SEM images of (a) untreated and treated cotton fabrics with ZnO nanoparticles after immersion into the reactional solution at (b) 0 h, (c) 1 h and (d) 2 h of reaction; ($b_1$, $c_1$, and $d_1$) zoomed images of the regions of the cotton fibers highlighted in (b), (c), and (d).

on the surfaces of the cotton fibers compared to those formed with an aging time of 2 h. Although it is difficult to know exactly the growth mechanism of the ZnO nanostructures on the surface of the cotton fibers, based on the XRD, SEM, and TEM results, a possible reason for this difference was concluded and is explained as follows.

The formation mechanism of crystals in solution mainly contains the formation of growth units and the incorporation of growth units into crystal lattice of the ZnO nanocrystals [35]. At the beginning of our process, $Zn(OH)_2$ clusters begin to develop immediately when a few drops of the $Zn(NO_3)_2{\cdot}6H_2O$ solution (at room temperature) are added into the reactor containing the heated KOH solution in higher basicity, which then turns turbid. This change in the color of the solution is an indication of the formation of $Zn(OH)_2$ clusters in the reactional medium. Then, these clusters will dissolve in $Zn^{2+}$ and $OH^-$ ions due to plenty of $OH^-$ ions in the solution to form $Zn(OH)_4{}^{2-}$ complexes [36–38]. These complexes will act in part as the basic growth units of ZnO nuclei by the dehydration of $OH^-$ ions [35]. The concentration of growth units in the solution increases until the critical supersaturation level is reached resulting on nucleation [39]. Therefore, the growth units become ZnO nuclei spontaneously in the aqueous solution.

After dripping of Zn precursor in the heated alkaline solution (aging time 0 h), the reaction medium looked milky white in color. At this time, the first sample of cotton fabric was dipped in this reactional medium. Once the cotton fabric

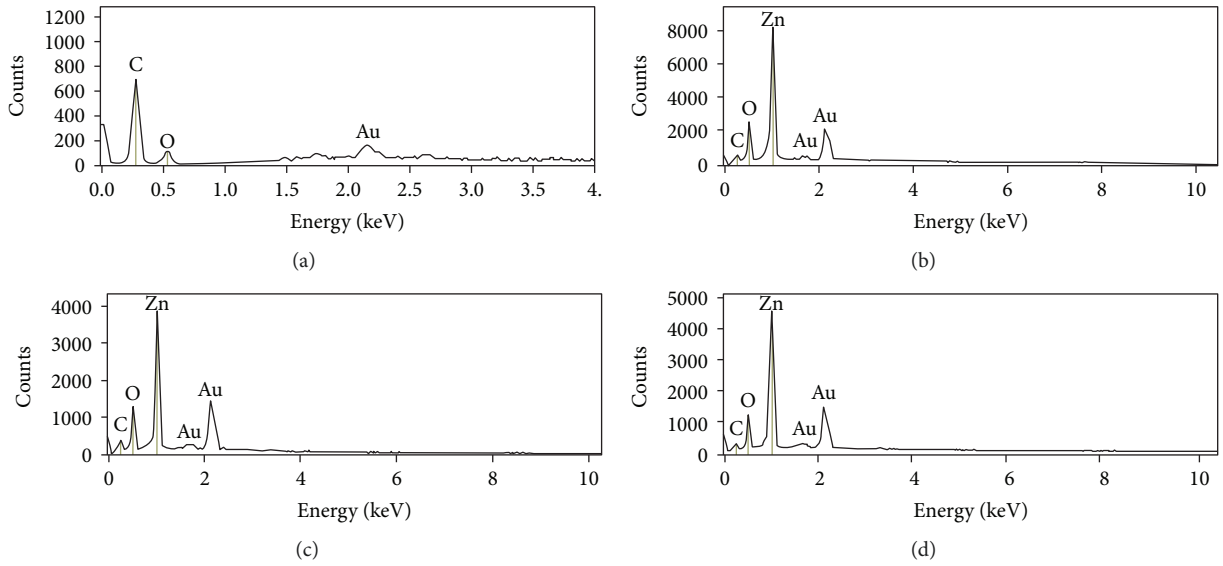


Figure 3: EDS spectra for the (a) untreated and treated cotton fabrics after their immersion into the reactional solution at (b) 0 h, (c) 1 h, and (d) 2 h of reaction.

was immersed into the $Zn(NO_3)_2{\cdot}6H_2O$ and KOH mixed solution, possible active groups of the fabric reacted with ZnO nuclei present in solution to readily adhere them on the surface of the fabric. At the beginning of the reaction (aging time of 0 h), the amount of growth units in the reaction medium not converted to ZnO nuclei may be large. As a consequence, the immersion of the fabric at 0 h resulted in a coating of the textile fiber with apparently few ZnO structures, as observed in Figure 2(b), probably due to a small amount of ZnO nuclei in the reaction medium. A similar result was also observed for the fabric sample treated at an aging time of 1 h (Figure 2(c)). SEM images also show that some ZnO structures grown on the fibers of the cotton fabrics immersed in the solution after 1 h of reaction have fairly large sizes. This result indicates that the large amount of growth units in the solution may lead to a faster growth kinetic [35, 40] of the ZnO nuclei already adhered to the cotton fiber instead of generating new ZnO nuclei on the fiber. Therefore, the condition of a few ZnO nuclei and many growth units in the solution may be the reason for the formation of large ZnO structures on the fiber surface at shorter aging times of the reaction.

As the reaction proceeds (2 h of reaction), most of the growth units are converted to ZnO nuclei which tend to adhere in large quantities to the surface of the cotton fabric. As a result, the fabric sample obtained with this condition showed a coating that covered almost the entire surface of the cotton fibers (Figure 2(d)). In addition, the ZnO nanostructures are smaller (as also evidenced by XRD analysis) and more homogeneous than those formed at 0 h and 1 h of reaction. A probable reason for this result is due to the fact that at the end of the reaction, the amount of ZnO nuclei in the solochemical solution was predominant with a number of growth units insufficient for the growth process of ZnO nuclei adhered to the cotton fibers.

### 3.5. Antibacterial Activity

*3.5.1. Qualitative Method.* The antibacterial property of untreated and treated cotton fabrics was examined against both Gram-positive (Figure 5) and Gram-negative (Figure 6) bacteria using the disc diffusion method (zone of inhibition test). The results clearly indicated that the untreated cotton fabric samples did not show antibacterial property. On the other hand, an inhibition zone can be seen around the cotton samples treated with the ZnO nanostructures. The halo size of treated cotton fabrics after 0 h, 1 h, and 2 h of reaction is $1.2 \pm 1.0$ mm, $2.5 \pm 0.8$ mm, and $2.1 \pm 0.2$ mm against *S. aureus* while $0.8 \pm 0.4$ mm, $2.5 \pm 0.5$ mm, and $2.4 \pm 0.2$ mm against *P. aeruginosa*, respectively. Therefore, better inhibition results were obtained for the fabric samples treated in the solochemical solution with aging times of 1 h and 2 h.

*3.5.2. Quantitative Method.* The results of the quantitative antibacterial activity of the treated fabrics at different aging times (Figure 7) were obtained by counting the number of bacterial colonies. As we can see in Figure 7, counting the number of bacterial colonies carried out with two hours of incubation has a faster antibacterial effect when tested against the *S. aureus* than that obtained against *P. aeruginosa.*

However, the cotton fabric treated in the solochemical solution at an aging time of 1 h showed a faster bacterial inhibition for both Gram-negative and Gram-positive bacteria than the other cotton samples.

The cell enumeration of *S. aureus* from the contaminated treated fabrics shows a decrease of the initial cell count from 5.57 log $CFU{\cdot}cm^{-2}$ to 0.0 log $CFU{\cdot}cm^{-2}$ after 2 h of incubation for the fabric sample treated in the solution at an aging time of 1 h and after 5 h of incubation for the cotton samples treated in the solochemical reaction at aging times of 0 h and 2 h.

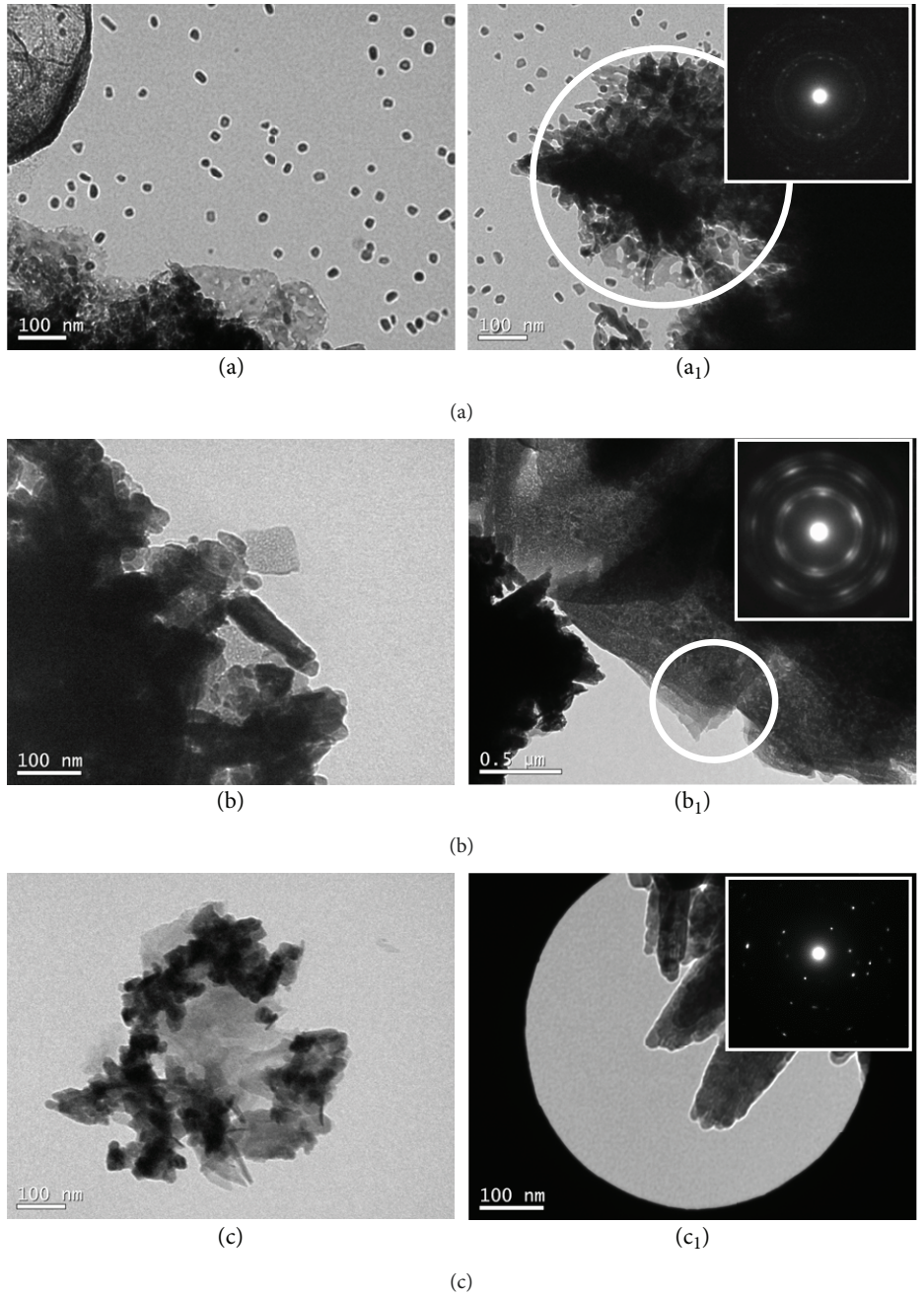


Figure 4: TEM images of ZnO nanostructures extracted from the cotton fabric samples immersed into the solochemical reactional medium after (a-$a_1$) 0 h, (b-$b_1$) 1 h, and (c-$c_1$) 2 h of reaction. ($a_1$, $b_1$, and $c_1$) TEM micrographs of ZnO nanostructures and their corresponding electron diffraction patterns (inset).

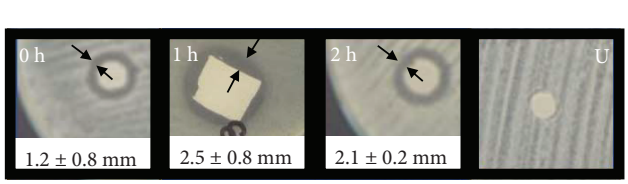


Figure 5: Zone of inhibition for the cotton fabric samples containing ZnO nanostructures against *S. aureus* after treatment at 0 h, 1 h, and 2 h of reaction. U represents the untreated cotton fabric.

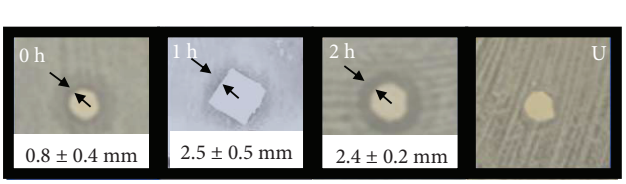


Figure 6: Zone of inhibition for the cotton fabric samples containing ZnO nanostructures against *P. aeruginosa* after treatment at 0 h, 1 h, and 2 h of reaction. U represents the untreated cotton fabric.

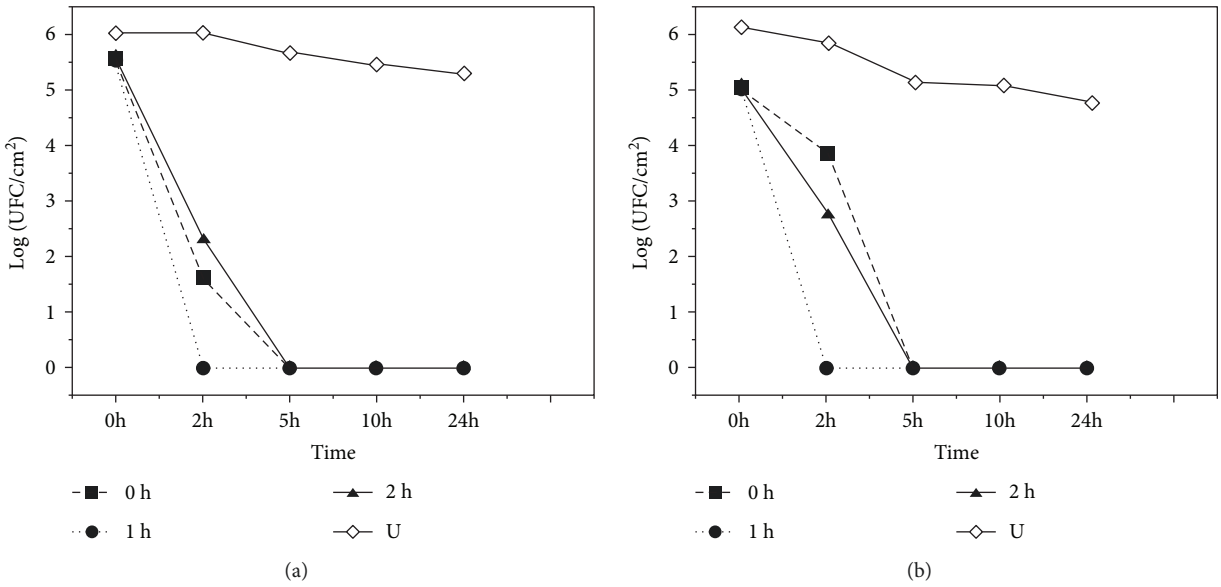


Figure 7: Time-kill curves of (a) *S. aureus* and (b) *P. aeruginosa* exposed to the cotton fabric samples treated in the solochemical reaction at aging times of 0 h, 1 h, and 2 h. U in (a) and (b) represents the untreated cotton fabric.

The results obtained for *P. aeruginosa* show a decrease of the initial cell count from 5.0 log CFU·cm$^{-2}$ to 0.0 log CFU·cm$^{-2}$ after 2 h of incubation for the cotton sample treated in the solution at an aging time of 1 h and after 5 h of incubation for the cotton fabrics treated in the solution with aging times of 0 h and 2 h.

The cell count performed on untreated cotton fabric shows a slight decrease of *S. aureus* and *P. aeruginosa* cell count from 6.03 log CFU·cm$^{-2}$ and 6.08 log CFU·cm$^{-2}$ to 5.30 log CFU·cm$^{-2}$ and 4.74 log CFU·cm$^{-2}$, respectively.

The results of quantitative antibacterial activity for a 2 h incubation time of the fabrics treated in the reactional solution with aging times of 0 h, 1 h, and 2 h show an antibacterial reduction of 23%, 100%, and 44% against *P. aeruginosa* and 71%, 100%, and 58% against *S. aureus*, respectively.

Thus, a strong antibacterial activity was observed against both *Staphylococcus aureus* and *Pseudomonas aeruginosa* for the fabric sample treated in the solochemical solution with an aging time of 1 h. For the other treated fabrics, higher activity was observed against *S. aureus* than *P. aeruginosa*.

## 4. Conclusion

A simple, rapid, and low-temperature solochemical technique has been successfully employed for the in situ growth of ZnO nanostructures on the surface of the cotton fabric. XRD analysis revealed the presence of hexagonal ZnO with a wurtzite structure on the treated cotton fabrics. The SEM photos showed that the ZnO nanostructures are well dispersed on the surface of the fibers, especially for the fabric sample treated after 2 h of reaction. Based on the XRD and TEM results, a possible *in situ* growth mechanism of ZnO nanostructures on cotton fiber was proposed. This study indicated that at the beginning of reaction, few ZnO nuclei and many growth units were available in the solochemical solution. When the fabrics were immersed in this solution at shorter aging times (0 h and 1 h), the ZnO nuclei immediately adhered to the surface of the fibers and the large amount of growth units around these nuclei led to a fast growth of ZnO particles. As a result, the ZnO nanostructures formed on these fabric samples were larger than those obtained after 2 h of reaction. The antibacterial activity results proved that ZnO nanostructures grown on the cotton fibers have potential to be used as an antimicrobial agent against *Staphylococcus aureus* and *Pseudomonas aeruginosa*.

## Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Acknowledgments

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