



In-Situ Deposition of Myco-Synthesized ZnO-NPs onto Cotton Fabrics for Durable Antibacterial Activity



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Abstract

ZnO-NPs have been recently recognized as a potent antibacterial agent used in medical textiles due to its activity against a broad spectrum of pathogenic bacteria. Herein, in situ deposition of ZnO-NPs onto cotton fabrics was formulated in a biological manner by the use of secreted fungal enzymes and proteins. Fungus *Pacelomyces* sp. AF-1 was harnessed in the preparation of ZnO-NPs via the use of its biomass filtrate as a medium for the deposition of ZnO-NPs onto cotton fabrics in the presence of zinc nitrate and NaOH as an alkali. Thorough chemical and physical investigations were conducted, including FTIR, XRD, TEM, UV-Vis spectra, Particle size, zeta potential and antibacterial activity. Results clearly revealed the successful deposition of ZnO-NPs onto cotton fabrics with size ranging from 5 to 20 nm with spherical well stabilized narrow size particles. By increasing the concentration of $Zn(NO_3)_2$ up to 1.0 mM, the size of the formed ZnO-NPs increased indicating the low stability of the formed ZnO-NPs. In addition, results also affirmed the antibacterial activity of cotton fabrics deposited ZnO-NPs against *E. coli* and *S. aureus* with high durability that kept their antibacterial activity after 15 washing cycles with bacterial reduction of about more than 50% which is considered as a highly durable antibacterial cotton fabric.

Keywords: Biosynthesis; ZnO-NPs; cotton fabrics; fungi; antibacterial;

1- Introduction

Nanotechnology is a young field of research that deals with altering the structure of matter at the atomic, molecular, and supramolecular levels between 1 and 100 nanometers in order to generate desired qualities and functionalities and for a variety of applications. The sustainable production of various nanoparticles (NPs) using cutting-edge methods has drawn the attention of the scientific community at large over the last few decades and has applications in the pharmaceutical, medical diagnostics and disease treatment, energy, electronics, agriculture, chemical, and space industries [1–5]. Researchers are now interested in synthesizing and producing

significant metal NPs utilizing more straightforward and effective processes thanks to the unique applications of metal nanoparticles in a variety of industrial fields [1, 2, 5]. Due to their drug delivery mechanism in cancer therapy, availability, material features, and ability to increase therapeutic selectivity against cancer cells, nanomaterials are being employed more frequently in medicine [6]. There are essentially two ways to create NPs. First, there is the chemical synthesis of NPs, which includes photochemical (using light photons) [7], physiochemical (using γ -radiation) [8], chemical reduction [9], micro-emulsion [10], electrochemical [11], microwave irradiation [12], and laser ablation [13, 14]. The biogenic strategy, in addition, enables us to recycle metallic salts that contaminate water supplies [15, 16]. NPs have a wide

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range of applications. In the field of biomedical science, NPs are frequently employed for imaging, drug delivery, rapid diagnosis, medical equipment, and tissue regeneration [17–18], textile industry [19], food product packaging [20], catalysis[21], cosmetic industry[22], sensor, [23], biological sciences [24], antimicrobial[25], optoelectronics [26], plasmonics [27],DNA sequencing [28], bioremediation and electronics[29]. Because it is less expensive and risky than chemical synthesis, it was decided to focus on bacterial, plant, and fungal biosynthesis. Zinc oxide (ZnO) with its exceptional photocatalytic activity, chemical stability during UV radiation exposure, thermal stability, and absorption of a broad spectrum of UV radiation [31, 32] can be widely applied in a variety of sectors. ZnO has received a lot of attention in the textile industry over the past ten years because to its strong UV blocking, antibacterial, photocatalytic self-cleaning, hydrophobic, and flame-retardant qualities when added to textiles [33–35].Ex situ and in situ synthesis are the two main chemical or biological processes used to produce ZnO-NP and apply it to textiles. The ex situ process consists of two steps: the first involves the synthesis of ZnO-NP as a separate reaction between a Zn-precursor and a precipitating/reducing agent, followed by calcination, and the second involves preparing the dispersion of the formed ZnO-NP in a separate bath and coating textiles with it [34].In situ synthesis is a one-step procedure in which the creation of nanoparticles and their simultaneous deposit on the substrate's surface are carried out [36–38].It results in a decrease in the number of processing steps and saves money, time, and energy. Additionally, ex situ is energy-intensive and unsuitable for in situ synthesis on textile surfaces since they are susceptible to such high temperatures. Very high calcination temperatures, such as those between 400 and 600 °C, are frequently used in synthesis [39–41].In general, it is anticipated that ZnO will crystallise when zinc salts are hydrolyzed in an alkaline solution[42].Consequently, the alkaline conditions are crucial for the production of ZnO-NP [36, 43].Low pH values are not ideal for insitu synthesis on cellulosic fabric because it is susceptible to the acidic medium, even if the pH value of the synthesis fluid considerably influences the characteristics, size, and morphology of the synthesisedZnO-NP [43–54]. In this research work, in situ biosynthesis of ZnO-NPs was performed onto cotton fabrics through employment of fungus biomass filtrate for imparting them a durable antibacterial activity. Thorough characteristic evaluations were conducted by the use of XRD, FTIR, UV-Vis., TEM, SEM, EDX, and antibacterial evaluation.

2. Materials and Methods

2.1. Materials

Cotton fabric (full bleached woven) was kindly provided from Miser El-Mehalla for weaving and Spinning Company, Egypt. All chemicals include Zinc nitrate $Zn(NO_3)_2$, ammonia (NH_4OH) and sodium hydroxide (NaOH) were obtained from SigmaAldrich Company.

2.1.1. Tested fungi

Pacelomycesp AF-1 strain was provided from the Regional Center for Mycology and Biotechnology, AL-Azher University, Nasr city, Cairo. The fungus was maintained on potato–dextrose Agar (PDA) slants.

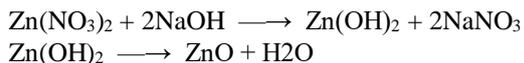
2.2. Methods

2.2.1. Preparation of mycelial biomass filtrate

Pacelomyces Sp. AF-1 was obtained through growing up in 250 ml Erlenmeyer Flask contains fermentative broth medium (100 mL, Czapek's Dox) at pH 6.0 for 3 days at 38 ± 2 °C.Then, the mycelial biomass was filtered by Whatman filter paper No. 1 followed by extensive wash for three times with sterile dis. water to remove the residual impurities. The washed mycelial mat of about 10g was resuspended in 100 ml sterile dis. water for 72 h at $38 \pm$ °C. followed by filtration to obtain the cell free filtrate which nominated as biomass filtrate.

2.2.2. In-situ preparation of ZnO-NPs onto cotton fabrics

The biomass filtrate of fungus Pacelomyces Sp. AF-1 was used in the preparation of ZnO-NPs onto cotton fabrics. Briefly, the obtained biomass filtrate (100 mL) was mixed, separately, with different concentrations of $Zn(NO_3)_2$ at room temperature. Typically, 0.5mM–2.5 mM of $Zn(NO_3)_2$ was used to study potential activity of biomass filtrate on the ZnO-NPs preparation. To the latter solution, cotton fabric (30 Cm x 30 Cm) was soaked in mixture of biomass filtrate and $Zn(NO_3)_2$ with gentle stirring for 30 min. following by adding of NaOH (0.5 %) drop-wisely until the white precipitate appears and then kept at room temp. for 30 min for complete deposition of ZnO-NPs onto cotton fabrics. At the end, the cotton fabric was eliminated and squeezed using squeezer machine to obtain final 100 % pick up and then allowed to dry at 80 °C. for 2 h.



2.3. Characterization of in situ bio-synthesized ZnO-NPs onto cotton fabrics

After decantation of the in situ treated cotton fabrics with ZnO-NPs, the remained solution was used for investigating the ex situ prepared ZnO-NPs using UV-Vis spectroscopy and Transmission Electron Microscopy (TEM).

UV-Vis spectroscopy

The preparation of nanoparticles is routinely monitored through using UV-Vis spectroscopy (JENWAY 6305 Spectrophotometer) where the relative Surface Plasmon Resonance (SPR) of ZnO-NPs can be clearly observed at wavelength (λ_{max}) 280 nm.

Transmission Electron Microscopy (TEM):

Size and shape of nanoparticles are often studied through morphological characterization of ex situ formed ZnO-NPs using TEM which gives clear information about both particles morphology as well as their size. (TEM, JEM-1230[JEOL, Japan]).

Particle size and zeta potential

The main particle size average and their surface charges (zeta potential) of the ex situ prepared ZnO-NPs were determined using Zetasizer Nano ZS (Malvern Instruments Ltd., GB). The stability of the formed nanoparticles can be also determined through study the surface zeta potential, which the agglomeration of particles occurs when the surface charges below +25 eV or -25 eV. For hgg deposition of ZnO-NPs onto cotton fabrics, following are the main analysis used.

Fourier Transform Infrared (FTIR)

Chemical changes on the surface of cotton fabrics after ZnO-NPs loaded was investigated via using Field emission scanning electron microscopy (ATR-FTIR, JASCO FT-IR 4100 spectrometer) over a wide range of 4000–400 cm^{-1} .

X-Ray Diffraction (XRD):

Crystallographic profile of ZnO-NPs was also analyzed by employing of X-Ray Diffraction patterns (XRD- 6000 series, Philips, Eindhoven, Netherlands)

Scanning Electron Microscopy:

Surface morphology of the in situ loaded cotton fabrics was characterized using SEM (SEM a Philips XL30).

Energy Disperse X-ray spectroscopy (EDX):

Further investigation into the chemical composition of the deposited ZnO-NPs was determined through EDX (a Philips-EDAX/DX4 energy-dispersive spectroscopy (EDS)).

2.4. Antibacterial activity of ZnO-NPs treated cotton fabric:

Antibacterial activity of cotton fabrics was qualitatively and quantitatively evaluated Against different microorganisms represent by *Escherichia coli* as gram-negative Bacteria and *Staphylococcus aureus* as gram –positive bacteria based on standard methods (AATCC 147 – 2004) and (AATCC test method 100 – 2004), respectively.

The antimicrobial treatment efficacy was determined by comparing the reduction in a bacterial colony (R %) of the in situ treated cotton fabrics with ZnO-NPs.

Washing fastness

In addition, washing fastness was also determined to investigate the durability of the treated cotton fabrics against the aforementioned bacteria through exposing to different washing cycles (5, 10 and 20 washing cycles) according to ISO 6330:2021. Bacterial activity was evaluated after 24 h of the incubation period and the percent reduction of Bacteria was calculated using the following equation:

$$R (\%) = [(A - B) / A] \times 100,$$

where R= the reduction rate, A = the number of bacterial colonies from untreated fabric, and B = the numbers of bacterial colonies from treated fabric. Negative controls (cotton fabric without loading with ZnONPs) were also run along with the experiment.

3. Results and Discussion

Biomass filtrate of *Pacelomyces* AF-1 was obtained by resuspending of the Harvested fungus mat (10 g) in 100 ml distilled water for 72 h at $\pm 28^\circ\text{C}$. Followed by decantation, the cell free biomass filtrate was used for preparation of ZnO-NPs in situ cotton fabrics.

In situ preparation of ZnO-NPs onto cotton fabrics

The biomass filtrate of *Pacelomycesp* AF-1 was used in the preparation of ZnO-NPs as following: Different concentration of $Zn(NO_3)_2$ were applied in order to study the Efficacy of biomass filtrates on the stabilization of ZnO-NPs. Cotton fabrics were soaked in the $Zn(NO_3)_2$ solutions with constant diameters (30 cm x 30cm) for 30 min with gentle stirring. Then NaOH was added drop –wisely to the latter mixtures for 30 min. White precipitates were appeared in these mixtures which indicate theinsituformation of ZnO-NPs onto the cotton Fabric as well as ex situ on the rest solutions. The cotton fabrics were then Collected and squeezed prior drying at 80 °C for 2 h. whilst, the remained Precipitates were also allowed to dry after collected by centrifuge at 80 °C for 2h. Both cotton fabrics and white precipitates were characterized to verify the preparation of ZnO-NPs

V-Vis spectroscopy

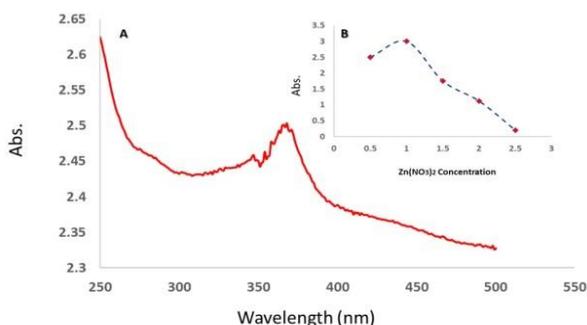


Figure 1: UV-Vis spectroscopy of ZnO-NPs prepared from biomass filtrate of fungus (A), effect of $Zn(NO_3)_2$ concentrations on the formation of ZnO-NPs (B)

UV-Vis spectroscopy as represented in figure 1 indicates the biosynthesis ZnO-NPs using biomass filtrate of fungus (*Pacelomyces* AF1). Results obtained from figure 1A, revealed that, the maximum absorption peak Observed from ZnO-NPs was located at wavelength 390 nm, which assigned to the Surface Plasmon resonance of ZnO-NPs. Therefore, the absorption band from figure 1 affirmed the successful formation of ZnO-NPs with polydispersed narrow size particles. 3Mm) on the ZnO-NPs formation. It can be seen from figure 1B that, the absorptions increase by increasing the Concentration of $Zn(NO_3)_2$ up to 1.0 mM, which indicates the ability of biomass Filtrate to well stabilizing the ZnO-NPs with spherical narrow sizes at 1.0 mM of $Zn(NO_3)_2$. Further increase in $Zn(NO_3)_2$ concentrations, the absorption decrease gradually to reach the low value at conc. 3.0 mM $Zn(NO_3)_2$. This

clearly indicates, the low ability of biomass filtrate to stabilize the formed ZnO-NPs at high concentrations exceeds mM $Zn(NO_3)_2$. From agglomeration and aggregation to large sizes. Hence, the optimal concentration of ZnO-NPs preparation was 1.0 mM $Zn(NO_3)_2$.

Transmission electron microscope (TEM)

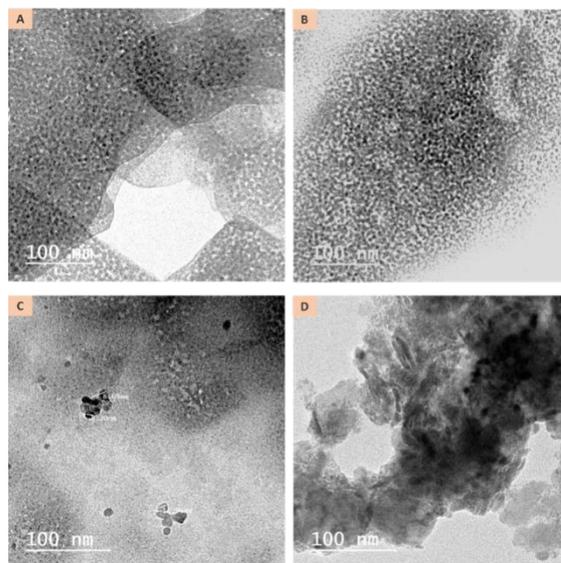


Figure 2: TEM of ZnO-NPs prepared from biomass filtrate of fungus at different $Zn(NO_3)_2$ concentrations; 0.5 mM (A), 1.0 mM (B), 1.5 mM (C) and 2.5 mM (D)

ZnO-NPs morphology and particle size were monitored by TEM as represented in figure 2. As clearly shown from figure 2, ZnO-NPs were successfully prepared with well dispersed narrow sizes ranged from 5 to 20 nm. Also, the shape of ZnO-NPs was varied regarded to the concentration Of $Zn(NO_3)_2$ used in the preparation, which the spherical shape was observed at concentration of $Zn(NO_3)_2$ up to 1.0 mM. By increasing $Zn(NO_3)_2$ concentrations, ZnO-NPs were formed with large aggregates and the heterogeneous shapes were also observed. These results are in agreement with that obtained from UV-Vis Spectroscopy.

Particle size and Zeta Potential

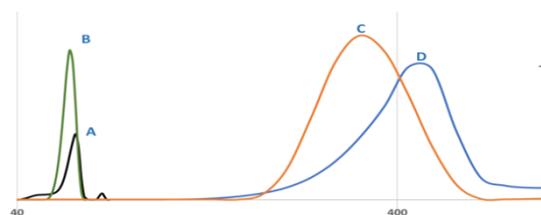


Figure 3: Particle size distribution of ZnO-NPs prepared from biomass filtrate of fungus at different $Zn(NO_3)_2$ concentrations; 0.5 mM (A), 1.0 mM (B), 1.5 mM (C) and 2.5 mM (D)

The particle size distribution and surface charge were routinely measured using DLS technique. therefore, figure 3 displays the particles size distribution of ZnO-NPs as well as this surface charge results from figure 3 depicted that, the ZnO-NPs were successfully prepared with narrow distributed size ranged from 50 to 70 nm as clearly appeared from figure 3 for low $Zn(NO_3)_2$ concentrations (Fig. 3A and 3B) with surface charge zeta potential ranged around from -38 to -42 eV, which due to the presence of fungus secretions (Proteins and enzymes) which proves, in turn, their role in stabilization of the formed ZnO-NPs. By increasing $Zn(NO_3)_2$ concentration up to 1.0 mM, the size of ZnO-NPs appeared in agglomerates which indicates lower stability due to decrease the affinity of the secreted fungal enzymes at the higher concentrations (Fig. 3C and 3D). This accumulation in ZnO-NPs size is accompanied with lower zeta potential which reported at -26 eV and -26 eV revealing the low stability of the formed nanoparticles.

Fourier Transform-Infra Red (FT-IR) spectroscopy.

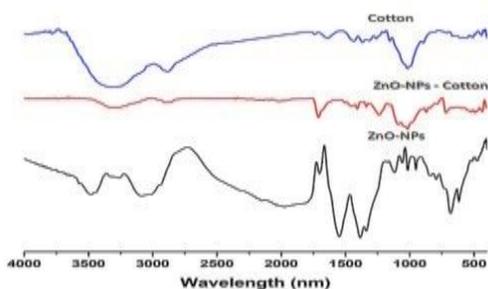
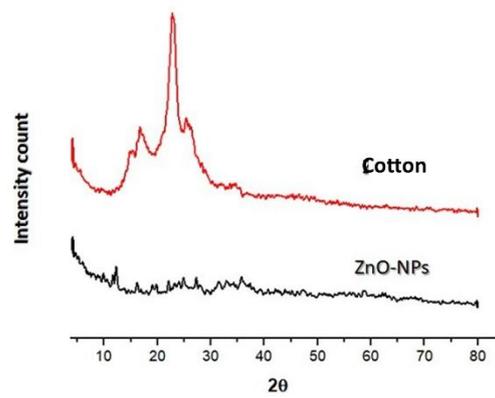


Figure 4: FTIR of the in situ biosynthesized ZnO-NPs onto cotton fabrics

FTIR represented in figure 4 showed a typical FT-IR for cotton and ZnO-NPs. Cotton IR showed a maximum absorption peaks 1500 cm^{-1} and 3600 cm^{-1} which are assigned to stretching vibration of C=O and O-H groups, respectively. Comparing to IR spectra of ZnO-NPs, new spectrum peaks are seen at near 512 cm^{-1} , which, indeed, is the characteristic peak of Zn-o absorption band. A weak band observed at 1025 cm^{-1} was attributed to C=O of the amino acid

stretching vibration. The blue shift in absorption at region 3420 cm^{-1} is may be due to the overlapping occurred between O-H of cotton and N-H of fungus proteins. A slight shifting in the absorbance of the amide groups (C=O) to 1544 cm^{-1} assert the formation of coordinating bond between fungus secreted protein and ZnO-NPs.

XRD analysis



XRD of the in situ biosynthesized ZnO-NPs onto cotton fabrics

Figure 5 represents the XRD of ZnO nanoparticles. From XRD, it can be determined the main sharp peaks at $2\theta = 31.5^\circ, 34.6^\circ, 36.4^\circ, 47.4^\circ, 56.5^\circ, 62.8^\circ, 66.1^\circ, 67.9^\circ$ and 68.9° , which are corresponding to crystal planes of ZnO-NPs (100), (002), (101), (102), (110), (103) and (112). These results declared that, the formed ZnO-NPs are polycrystalline wurtzite structure (Zincite, JCPDS 5-0664) (56).

SEM and EDX

The surface of the loaded cotton fabrics was investigated by the insitu deposition of ZnO-NPs using SEM as shown in Figure 6. It is evident from Figure 6 that the ZnO-NPs were successfully deposited on the surface of the cotton fabrics with homogeneous distribution over the domain area effectively as compared to the blank cotton fabrics which exhibited a soft appearance.

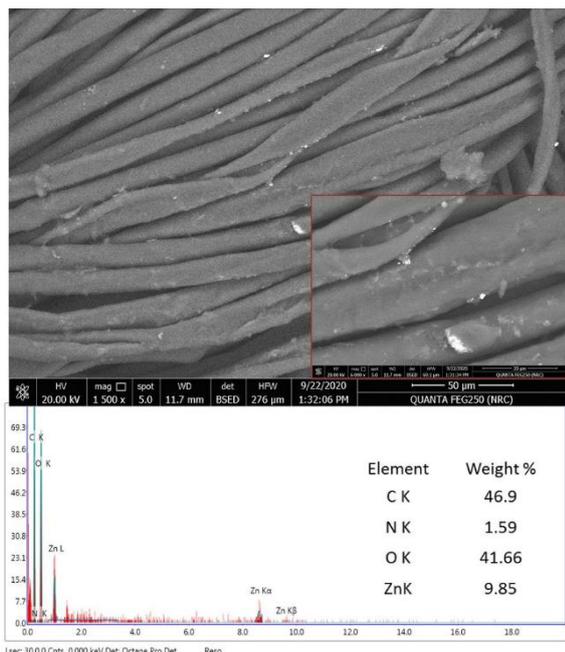


Figure 6: SEM of the in situ biosynthesized ZnO-NPs onto cotton fabrics

Moreover, EDX analysis manifested the presence of ZnO-NPs as indicated from the chemical compositions of the treated cotton fabrics with about 10% of the total weight %.

Antibacterial activity of ZnO-NPs

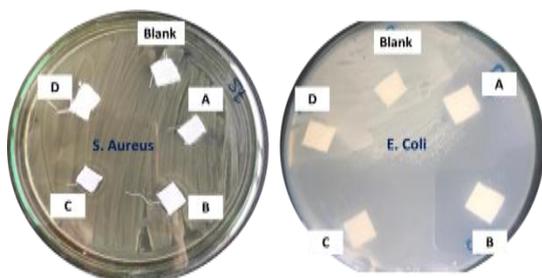


Figure 7: Antibacterial activity of in situ synthesized ZnO-NPs cotton fabrics against *E.coli* (right plate) and *S. aureus* (left plate) at different $Zn(NO_3)_2$ concentrations; A – D for 0.5 mM, 1 mM, 1.5 mM and 2 mM, respectively.

The main importance of ZnO-NPs is their biological activity as antibacterial agent against wide range of (pathogenic) harmful bacteria. Thus, the antibacterial activity of the as biosynthesized ZnO-NPs was

examined against gram positive and Gram-Negative bacteria as represented in figure 7.

Figure 7 showed, obviously, the potent antibacterial activity of cotton fabrics treated with insitu biosynthesized ZnO-NPs against both *E. coli* and *S.aureus* using the inhibition Zone method as a qualitative way to visualize the inhibition activity of cotton treated fabrics. Further, the clear Zone indicated from figure 8 revealed the high antibacterial activity of the in situ prepared ZnO-NPs onto cotton fabrics on gram Negative *E.coli* bacteria than gram positive *S.aureus* bacteria, furthermore, the antibacterial activity increased with increasing the concentration of $Zn(NO_3)_2$ up to 1.5 mM . upon increase in $Zn(NO_3)_2$ concentration , the diameter of clear zone decrease which indicating the limited antibacterial activity against both pathogenics *E.coli* and *S.aureus* resulting from low affinity of ZnO-NPs as previously emphasized by UV-Vis.spectroscopy at high $Zn(NO_3)_2$ concentration.

Washing Fastness

Table 1: Antibacterial activity as a function of bacterial reduction %of the in situ biosynthesized ZnO-NPs onto cotton fabrics after different washing cycles.

Bacterial Reduction %		
Washing cycles	<i>E. coli</i>	<i>S. aureus</i>
0	98	96
5	76	72
10	64	60
15	54	51

Results obtained from table 1 announced that, the bacterial reduction of the in situ treated cotton fabrics with ZnO-NPs attained the 98% and 96% for *E.coli* and *S.aureus* , respectively , which indicated the powerful bacterial activity cotton fabrics treated ZnO-NPs against gram Negative ,Gram positive bacteria.in addition to this, the antibacterial activity of cotton fabrics was significantly affected by the washing cycles. The reduction of bacterial activity decreased gradually by increasing the washing cycles numbers, which attains the 50% after 15 washing

cycles. These results confirm the effective route used for insitu preparation of ZnO-NPs into cotton fabrics, which as antibacterial activity reserved up to 15 washing cycles with more than 50% bacterial reduction activity. These results are in the agreement with the observation concluded from figure 8, which the loaded cotton started to become smoother with increasing wash cycles. This may be due to the release of physically bounded during deposition of ZnO-NPs onto cotton fabrics. After an extensive washing upto 15 cycles, the chemically bounded particles were forced to release to reduce the antibacterial activity by 50%.

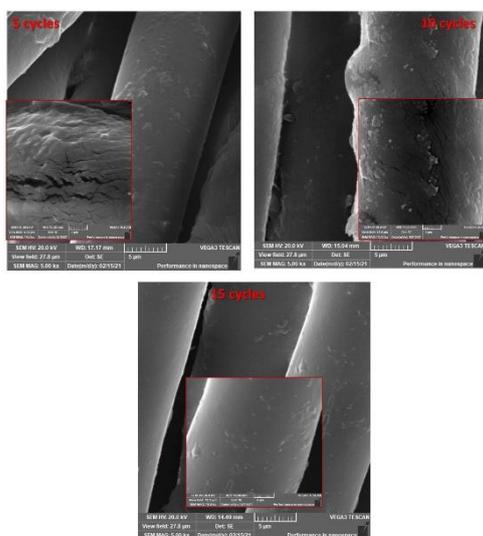


Figure 8: SEM of the in situ prepared ZnO-NPs onto cotton fabrics in the function of differnt washing cycles

4. Conclusion

Herein, ZnO-NPs were biosynthesized through in situ mechanism inside the cotton fabrics to reach high durability with high performance by the use of fungal cell filtrate. *Pacelomyces* sp. AF-1 strain was harnessed successfully for fabricating of ZnO-NPs with well stabilized by using $Zn(NO_3)_2$ at different concentrations through investigating the resultant ZnO-NPs in situ cotton fabrics and ex situ for UV-Vis, TEM and Zeta protentional assessments. Results declared the preparation of ZnO-NPs with size ranged from 5-20 nm at the optimal condition of 1.0mM $Zn(NO_3)_2$. FTIR and XRD also affirmed the presence of ZnO-NPs in chemical connection with cotton fabrics as ZnO reduced to nano-size during precipitation from solution and accumulated onto cotton fabrics by the dint of biomass filtrate of fungus *Pacelomyces* sp. AF-1 strain. Moreover, the

antibacterial activity of in situ biosynthesized ZnO-NPs onto cotton fabrics was achieved through investigating their activity towards the most common pathogenic bacteria, viz., *E. coli* and *S. aureus*, where the activity attained the maxima in case of *E. coli* due to its high sensitivity towards ZnO-NPs, where the bacterial reduction % reached 98% and 96%, respectively for *E. Coli* and *S. aureus*. Durability was the cornerstone of our story that the results revealed the effective manner of the treatment of cotton fabrics with in situ synthesized ZnO-NPs for rendering them more durable with satisfied antibacterial activity of up to 15 washing cycles with bacterial reduction of more than 50%.

Conflicts of interest

“There are no conflicts to declare”.

5. References

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