FABRICATION OF MULTIFUNCTIONAL NANO GELATIN/ZINC OXIDE COMPOSITE FIBERS

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Abstract:

*According to health studies, reinforcing gelatin is necessary in order to obtain the multifunctional material. In this study, nano zinc oxide (ZnO; at concentrations of 0.5%, 1% and 1.5%) was doped with gelatin and the solution was electrospun under specific conditions to obtain multifunctional gelatin/ZnO nanofibers. The morphology of the nanofibers was studied by field emission scanning electron microscope (FESEM), and energy-dispersive X-ray spectrometry (EDX) analysis indicated the presence of nano Zn on the surface of gelatin fibers. On the contrary, elemental mapping analysis proved the distribution of nano material along the nano gelatin fibers. The results show that the produced nano gelatin/ZnO composite increases the ultraviolet (UV) blocking of fabric significantly. It is also observed that electrospun gelatin/ZnO nanofibers have excellent bactericidal property against both *Bacillus cereus* (Gram-positive) and *Escherichia coli* (Gram-negative) bacteria.*

Keywords:

Gelatin, zinc oxide, electrospinning, bactericide, UV blocking

1. Introduction

Gelatin or gelatin is a semitransparent biomaterial which is extracted from the collagen of the skin, bones, and connective tissues (one of the four basic types of animal tissues) [1–3]. The hydrolysis of collagen produces a water soluble protein which is breakable in dry conditions and resinous when moist [4, 5]. The protein is also soluble in most polar solvents. Gelatin is frequently applied as a gelling material in the food industry, in medications, beauty products, and makeup items, because of its competencies like biocompatibility, biodegradability, and commercial accessibility. Due to the hydrolysis of collagen, gelatin acquires similar amino acids such as proline (~25%), glycine (~20%), glutamic acid (~11%), arginine (~8%), alanine (~8%), and other essential/non-essential amino acids. It must be noted that glycine and arginine are the two forerunner amino acids for biosynthesis of creatine [6–10].

In the fields of fiber science, textiles, and polymers, gelatin is simply formed into hydrogels, films, and fibers, although there are not too many published papers on gelatin nanofibers and gelatin nanotextiles. Kwak et al. [2] reported that by wet spinning of gelatin fiber and gelatin/Ag fiber, its antibacterial property grows. The innovation in our work is using the electrospinning method for fabrication of gelatin fiber in order to obtain nanofibers. However, in this work we did not use Ag, so that the final product does not have cellular toxicity (research works have proved the cellular toxicity of Ag particles [11]). One of the simple methods for producing nanofibers is by electrospinning.

The electrospinning process comprises a polymer solution which is charged, and by overcoming the surface tension, a thin layer of web polymer is formed on the surface by ejection of polymer solution, which gives rise to elongation of the polymer and formed ultrathin fiber [12–17].

Electrospinning nanofibers have many medical applications such as wound dressing [18], drug delivery, and dental restoration, as well as engineering applications such as filtration and tissue. The poor mechanical properties of these fibers (like Young's modulus) can be enhanced by combining them with carbonaceous nanomaterials [19–22].

Recently, nanotechnology and nanomaterials are being used in a wide area of fabric formation. Using these nanomaterials gives novel specifications to the final produced textile. In the past decade, scientists have focused more on using semiconductors (in the scale of nanometers) such as nano CdS, nano Fe₂O₃, nano TiO₂, nano Ce, and nano ZnO [23–28].

Nano ZnO is one of the semiconductors which have an energy band gap of 3.3 eV. This semiconductor has many utilizations such as ultraviolet (UV) persistence, bactericide, photocatalytic activity, and low toxicity. Nano ZnO is a nanomaterial that can be sedimented on textile surfaces using ultrasonic devices. The energy of ultrasonic irradiation can have remnant nanoparticles on the surface of fibers with minimum aggregation [29–32].

In this study, nano gelatin/ZnO composite has been electrospun and then studied for its antibacterial and UV-blocking properties.

2. Materials and methods

White to peel yellow gelatin powder from Sigma Aldrich, acetic acid from Merck, and nano ZnO from Sigma Aldrich were purchased. An amount of 1 g gelatin was dissolved in a solution of 30/70 mL acetic acid/distilled water (respectively) and sonicated for 30 min. The reason for using acetic acid is previous works have concluded that using acetic acid can reduce the diameter of electrospun fibers [33, 34]. In addition, Choktaweessap et al. [35] report that acetic acid creates electrical conductivity, and thus the electrospinning is done in a better condition. During this process, nano ZnO solution was prepared in different concentration (0.5%, 1%, and 1.5%). Then, nano ZnO solution was added to gelatin and sonicated again for 60 min in order to distribute the nanomaterial and to prevent aggregation. After obtaining a homogeneous solution, it was loaded into a syringe (3 mL, with blunt needle), which was placed in the device setup. The nanofibers of gelatin/ZnO were electrospun. The device voltage was 20 kV, pumping rate 0.3 mL/h, collector-needle distance 13 cm, and drum speed 300 rpm. The collector was coated with aluminum sheet.

Nanofibers of gelatin/ZnO morphology were studied using field emission scanning electron microscope (FESEM) (MIRA3-TESCAN) and a Euronda ultrasonic bath model (Eurosonic 4D), 350 W, 50/60 Hz (Italy). Perkin Elmer Lambda UV-vis spectrophotometer was used to study the UV-blocking properties of the samples.

The method and condition of bactericidal property was done through AATCC 100-2004 against both *Bacillus cereus* (Gram-positive) and *Escherichia coli* (Lab Assistant Summer Internship

Gram-negative) bacteria. The bactericidal property was calculated as follows:

$$\text{The reduction of bacteria (\%)} = \frac{(A-B)}{A} \times 100 \quad (1)$$

where A is the number of bacteria recovered from the inoculated treated test specimen incubated over 24 h while B

is the number of bacteria recovered from the inoculated treated test specimen immediately after inoculation.

3. Results and discussion

3.1. FESEM, map images, and EDX analysis

In order to study the morphology of electrospun nanofibers, FESEM images were obtained. FESEM was done at 15 kV and 100 kx magnification. Figure 1(B) clearly demonstrates the presence of nano ZnO and its distribution. As shown, the average diameter of nano gelatin fibers is 40 nm and the average particle size of nano ZnO is 25 nm. The figure also shows a good distribution of nanoparticles without any aggregation or agglomeration. This proves to be a suitable method and condition for preparing the solution and electrospinning. Figure 1(A) indicates that nanofibers of gelatin (without nano ZnO) were electrospun correctly with a minimum diameter of approximately 11 nm for these fibers. Figure 1(B) shows the distribution of nano ZnO particles, and as shown in red, the particle size ranges between 21 and 31 nm.

For the chemical characterization or elemental analysis of materials of the finished samples, energy-dispersive X-ray spectrometry (EDX) was done. The energy peaks correspond to the various elements in the sample. The main element detected through EDX on the treated sample is Zn, which is related to the use of nano ZnO.

On the basis of EDX analysis (Figure 2), it is observed that the treated sample contains a significant amount of nano ZnO, which indicates the presence of Zn on the fiber surface.

In order to demonstrate and prove the distribution of nano Zn on electrospun gelatin nanofibers, elemental mapping analysis was done. Figure 1(C) indicates that nano Zn particles have great distribution along the fibers.

Figure 1. FESEM images of (A) blank sample, (B) gelatin/ZnO, and (c) elemental mapping of gelatin/ZnO.

Figure 2. EDX image of gelatin/ZnO sample.

3.2. UV blocking analysis

The UV transmission diagram of electrospun gelatin nanofiber containing different amounts of nano ZnO is illustrated in Figure 3. The irradiation wavelength was 200–400 nm. As it indicates, the electrospun gelatin nanofiber without any nano ZnO (blank sample) has a higher transmittance compared with those containing nano ZnO. That is, UV protection of the blank sample is low. Moreover, the UV-blocking activity of the electrospun fiber is improved in the presence of nano ZnO, which is owing to the synergetic UV absorption of nano ZnO. Also, the spectrum shows that the sample containing 1.5% nano ZnO has greater protection against UV radiation, so by increasing nano ZnO in gelatin nanofiber, the UV blocking of samples is enhanced.

3.3. Bactericidal properties detection

Bacillus cereus and *E.coli* are two common Gram-positive and Gram-negative bacteria. *Bacillus cereus* is a rod-shaped bacteria which is found in abundance in food and soil and can cause nausea, diarrhea, and vomiting syndrome [36]. While the Gram-negative bacteria *E.coli* can cause gastroenteritis, urinary tract infections, neonatal meningitis, hemorrhagic colitis, and Crohn's diseases [37]. Nanoparticles of ZnO have unparalleled specifications such as photocatalytic property and bacteriostatic effects [38]. Furthermore, nano ZnO is biocompatible for pharmaceutical textile usages [39]. Nano ZnO can prevent the increase of bacterial cells by penetrating the hydrogen peroxide into the cell membranes and causing destruction of the bacteria. In addition, the positive ions of zinc (Zn^{2+}) can react with the negative particles of bacteria cell and thus lead to the death of bacteria. In this study, these two kinds of bacteria have been used to investigate the antibacterial efficiency of electrospun nano gelatin/ZnO. The results of the antibacterial experiment are presented in Figure 4. As it is demonstrated, there is no antibacterial property on the blank sample against both Gram negative and positive bacteria, but the samples that contain nano ZnO have antibacterial property. As shown in Figure 4, the sample containing 1.5% nano ZnO has 100% antibacterial property against both the bacteria. As it is shown, the antibacterial activity of the sample treated with

Figure 3. UV transmittance diagram of electrospun samples.

	<i>Bacillus cereus</i>	<i>E.coli</i>
Blank Sample	0%	0%
0.5% nano ZnO	96.1%	97.4%
1.0% nano ZnO	98.2%	100%
1.5% nano ZnO	100%	100%

Figure 4. Antibacterial efficiency of blank and treated samples.

1% nano ZnO is about 100% and 98.2% for *E. coli* and *Bacillus cereus*, respectively, and for the sample containing 0.5% nano ZnO, it is 97.4% and 96.1% respectively. Comparing the antibacterial property of samples, it is determined that the antibacterial property of samples against *E.coli* is more than that of *Bacillus cereus*, because of the difference in the thicknesses of the cell walls. *Bacillus cereus* has a thicker cell wall.

4. Conclusion

This study explains the successful fabrication of gelatin/ZnO nanofiber through electrospinning. Four gelatin samples (0.0%, 0.5%, 1%, and 1.5% ZnO) were electrospun in specified conditions. Elemental mapping analysis of samples from FESEM shows the excellent distribution of nano Zn on electrospun gelatin nanofibers. However, while the transmission spectrophotometer outcome shows good UV blocking of the nano gelatin/ZnO composite, the blank sample does not

have suitable UV blocking, but by doping nano ZnO, its UV blocking property enhances tremendously. This is because of the UV-blocking property of nano ZnO and its synergetic UV adsorption. Also, the bactericidal property of samples was investigated with both Gram negative and Gram positive bacteria, *E.coli* and *Bacillus cereus*. The results indicate that nanofibers of gelatin/ZnO have more than 96% antibacterial property against both bacteria.

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