

THE EFFECT OF ZnO NANO PARTICLE COATING AND FINISHING PROCESS ON THE ANTIBACTERIAL PROPERTY OF COTTON FABRICS

Farhana Momotaz*, Ayesha Siddika, Md. Tashrif Shaihan and Md. Anisul Islam

Department of Textile Engineering, Khulna University of Engineering & Technology, Khulna-9203, Bangladesh

Received: 06 February 2020

Accepted: 13 May 2020

ABSTRACT

*In this study, the ZnO nanoparticles solution was directly applied on to the 100% cotton knit & woven fabric to impart antibacterial property using both spin coater & pad-dry-cure method. Then, the disc diffusion method was used to assess the antibacterial activity of the finished fabrics. The topographical analysis of untreated, treated, and washed fabrics of different structures (knit and woven) were studied and compared. The results showed that the finished fabric demonstrated significant antibacterial activity against *S. aureus* and *E. coli*. As per result, it was also found that double jersey (rib) fabric showed highest amount of bacterial protection & pad-dry-cure method showed better result against bacterial attack than spin coating method. Moreover, gram negative bacteria (*E. coli*) showed more strength against antibacterial treated unwashed and washed fabrics than gram positive bacteria (*S. aureus*).*

Keywords: Antimicrobial property; Nano-particle; Pad-dry-cure method; Spin coater

1. INTRODUCTION

The nano-world is the intermediary between the atom and the solid, from the large molecule or the small solid object to the strong relationship between surface and volume (Nouailhat, 2008). The term nano originated from the Greek nanos which suggests 'dwarf'. It is one billionth of a meter (Gazit and Mitraki, 2007). Nanotechnology is the science that deals with matter at the scale of 1 billionth of a meter. It is also the study of manipulating matter at the molecular and atomic scale. Nanoparticle, the most fundamental component in the fabrication of a nanostructure is far smaller than the world of everyday objects, and is described by Newton's laws of motion. However, it is bigger than an atom or a simple molecule that are governed by quantum mechanics (Horikoshi and Serpone, 2013).

When conventional materials formed from nanoparticles, many of its properties changed. Nanoparticles are reactive and effective than other molecules because of having a greater surface area per weight than larger particles. Nowadays, a very promising scientific research is nanoparticle research because of the wide range of potential and promising applications especially in electronic fields, optical, and biomedical. This new concept changes the use of such material in microscale size to be used in absolutely new and advanced applications by using a nanoscale size of the same material (Hashim, 2012). The structure of the nanomaterials can be classified by their dimensions. The zero-dimensional nanostructures are called nanoparticles (Alagarasi, 2011). Recently, one-dimensional nanostructures with different morphologies (such as nanowires, nano-rods (NRs), and nanotubes) have become the focus of intensive research, because of their unique properties with potential applications. Among them, zinc oxide (ZnO) nanomaterials has been found to be highly attractive, because of the remarkable potential for applications in many different areas such as solar cells, sensors, piezoelectric devices, photodiode devices, sun screens, anti-reflection coatings, and photo catalysis (Hatamie *et al.*, 2015).

Zinc oxide nanoparticles (ZnO NPs), as one of the most important metal oxide nanoparticles, are popularly employed in various fields due to their peculiar physical and chemical properties (Smijns and Pavel, 2011) (Ruszkiewicz *et al.*, 2017). Zinc oxide nanoparticles (ZnO NPs) are used in an increasing number of industrial products such as rubber, paint, coating, and cosmetics (Jinhuan *et al.*, 2018). In addition, ZnO NPs have superior visible light resistance, antibacterial deodorant, antibacterial, antimicrobial, and excellent UV-blocking properties (Hatamie *et al.*, 2015).

Due to their potential for reducing the transmission of infection in medical and health care environments, antimicrobial textiles have attracted a great interest in recent years. In addition, in these fabrics, the formation of unpleasant odors is reduced. Therefore, it is possible to have a pleasant and fresh smell from those textiles still after use. Therefore, antimicrobial products have long been used in health care environments for hospital gowns, patient clothes, curtain, bed cover, infection control, wound healing, hygiene, etc. The effectiveness of antimicrobial action on textiles depends on the antimicrobial, the concentration, and the application method of the antimicrobial on the textile (Islam and Butola, 2019).

Microbes are very small lives on the earth which can't be seen by the naked eyes. They may be known as micro-organisms like bacteria, fungi, algae, and viruses, etc. These bacteria are sub classified into different groups

* Corresponding Author: farhanatex@yahoo.com;

<https://www2.kuet.ac.bd/JES/>

namely gram positive (*Staphylococcus aureus*), gram negative (*Escherichia coli*), spore bearing or non-spore bearing types. Apart from that some are pathogenic and cause cross infection bacteria whereas some microbes like fungi, molds or mildews are shown slower growth rate due to its complex nature (Textile News, 2019).

Various studies have already been done on zinc oxide nanoparticles as antibacterial substance by many researchers. The ability of the antibacterial agent to inhibit bacterial growth was first tested using a disc diffusion method (Singh *et al.*, 2012). ZnO nanoparticles treated fabric shows higher antibacterial activity when compared with ZnO bulk treated fabrics whereas the untreated fabrics showed no antibacterial activity (Rajendran *et al.*, 2010). It has also found that the ZnO nanoparticles constitute an effective antimicrobial agent against pathogenic microorganisms (Gunalan *et al.*, 2013). A zinc oxide nanoparticle is used as an antibacterial substance against *E. coli* and *S. aureus* and exhibits the highest toxicity against microorganisms. It has also been demonstrated from SEM and TEM images that zinc oxide nanoparticles first damage the bacterial cell wall, then penetrate, and finally accumulate in the cell membrane (Khwaja *et al.*, 2018).

In this study, two different methods have been used to apply the ZnO nanoparticles named spin coating and pad-dry-method. Spin coating is known as a procedure used to deposit uniform skinny films to flat substrates. In this coating system, very few amount of coating material is applied on the center of the substrate, which is either spinning at low speed or not spinning at all. The substrate is then rotated at high speed in order to spread the coating material by centrifugal force. A machine used for spin coating is called a spin coater, or simply spinner. Pad-Dry-Cure or Exhaust-Dry-Cure is a finishing process applied to textiles to impart different finish treatments, such as waterproofing, softening, antibacterial or anti-odor finishes. A water-based solution bath contains finishing chemicals through which the textile materials pass. The textile is then dried and cured using heat and pressure. This is known as the parent process for various finishing treatments. Then antibacterial analysis of the treated and untreated samples was carried out by disc diffusion method (Hossain *et al.*, 2014). To determine the effective ness of the finishing process and coating, the diameter of the zone of inhibition produced by the coated samples has been measured and compared with each other.

2. METHODOLOGY

2.1 Raw Materials

Three types of fabric i.e. single Jersey (100% cotton, GSM 180), double Jersey (100% cotton, 1X1 Rib), woven fabric (100% cotton, 1/1 plain weave) have been used for this research. ZnO nanoparticles (in powder form), acrylic binder, sodium lauryl sulphate, and soap have been used for the treatment.

2.2 Preparation of Fabric

At first, the grey fabrics were taken. Then scouring and bleaching were done in all fabrics with required recipe. This RFD fabric of woven, single jersey & double jersey were ready for following process.

2.3 Preparation of Nanoparticles Solution

2% ZnO solution & 1% acrylic binder solution were taken. Both mixed together with the help of stirrer. For fine and uniform mixing, the solution was stirred in ultra-sonicator at 30 °C temperature for 20 minutes.

2.4 Application of Nanoparticles Solution

In spin coating method, according to fabric weight, the amount of mixed ZnO & binder solution was applied on circular shaped fabric through one by one drop by using a syringe. The fabric was then rotated at 2000 rpm in order to spread the fluid by centrifugal force. Later, the fabric sample was cured at 140°C for 3 minutes.

In pad-dry-cure method, the mixed solution prepared by the ultra-sonicator was taken in a beaker. Then fabric samples were soaked in ZnO & acrylic binder mixed solution for 5 minutes. Later, the treated fabric samples were padded by using padder machine. Finally, samples were cured at 140°C for 3 min.

2.5 Washing of Samples

Both spin and pad-dry-cured fabrics were washed once with sodium lauryl sulphate 2 times to eliminate the unfix particles from the treated fabric samples. ISO C01 method was used for washing the samples. Some spin and pad-dry-cured samples were washed 3 times. Each time washing was done at 40°C for 30 minutes in washing machine by 0.5% soap according to ISO C01 test method. Later the fabrics were air dried.

2.6 Antibacterial Test

36 samples using 12 media plates were tested in disc diffusion method to find antibacterial activity. The sequence of working procedure is given below:

- Bacteria were sub-cultured from stock by keeping bacteria in incubator for overnight.
- Nutrient broth and agar media were prepared according to the requirements.
- Nutrient broth media were prepared at 2 conical flasks (20 ml/conical flask) & 300 ml agar media were prepared in a separate conical flask.
- The nutrient broth containing conical flasks were incubated for 24 hours.
- For serial dilution purpose 8 dewater test tubes (9 ml/test tube) were taken. All test tubes were closed by foil paper or cotton plug.
- 2 bacteria's colony was added to the nutrient broth containing 2 conical flasks media and incubation is done in it for 24 hours.
- After that, bacteria containing nutrient broth conical flasks were serial diluted up to 10^4 times.
- On 12 circular plates, firstly agar solution was added and when they turn in semi solid form, 1 ml of bacteria solution was added on each plate respectively & spread the solution with the help of spreader.
- In this way, three types (single jersey, double jersey/rib, woven) of fabrics were used. Untreated, treated, and 3 times washed fabrics of each type and each method were tested by observing and conditioning in incubator overnight at 37°C temperature.
- Finally, the result was observed by measuring the area around the fabric which is getting resistance from bacterial attack.

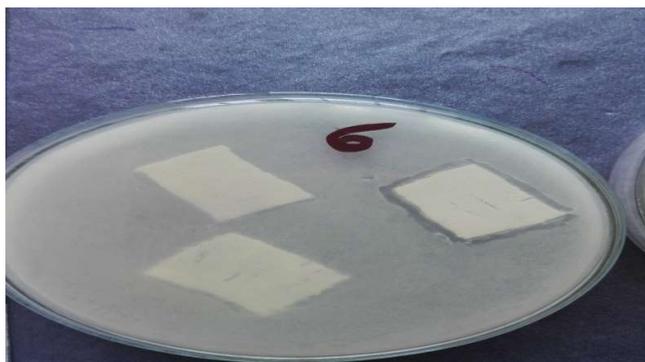


Figure 1: Sample fabrics after incubation overnight at 37°C temperature.

3. RESULTS AND DISCUSSION

Table 1 represents the results of three types of fabric (woven, single jersey, rib/double jersey) treated with ZnO NP by pad-dry-cure method and then applied by *S. aureus* and *E. coli* bacteria. It showed the antibacterial activity of untreated samples, and ZnO nanoparticle coated samples on *E. coli* and *S. aureus* bacteria respectively. Here, after overnight incubation the resisted area of eighteen samples is found where twelve samples: four woven, four single jersey and four double jersey were treated with ZnO nanoparticles in pad-dry-method whereas two woven, two single jersey and two double jersey were untreated samples. Among these twelve treated samples, six samples were given three times wash. The area of the sample fabric was 352 sq. mm for each. Antibacterial activity results showed that the ZnO nanoparticle has exhibited strong antibacterial activity against both gram-positive (*S. aureus*) and gram-negative (*E. coli*) bacteria. However, antibacterial activity of ZnO particles were greater on gram-positive than gram-negative bacteria. It can be assumed that the outer thick peptidoglycan layer and other surface components of gram-positive bacteria may promote ZnO attachment onto the cell wall whereas the components of gram-negative bacteria may repeal this attachment (Getie *et al.*, 2017). In addition, among three samples (woven, single jersey and double jersey/rib) it is found that the treated double jersey has greater antibacterial activity on gram-positive (*S. aureus*) bacteria. Generally, the gram per square meter (G.S.M) of double jersey fabric is higher than the single jersey fabric. It is assumed that, double jersey fabric absorbs high amount of ZnO NP from the solution than single jersey fabric which subsequently shows higher antibacterial ability. After overnight incubation there was a clear visible zone in which the bacteria could not survive and that zone was created around the fabric having an area of 720 sq. mm whereas the untreated double jersey showed zero resisted area. So, it can be said that the treated fabric without wash showed the best anti-biotic properties. And there was a significant reduction in anti-microbial property after three time washing and the non-treated fabric exhibits no resistance at all.

The comparison of the three types of fabric treated with pad-dry-cure method after applying *S. aureus* and *E. coli* bacteria are presented in Figure 2. From the chart it is clearly understood that the double jersey/rib fabric showed the maximum anti-bacterial property. And the anti-bacterial treatment resists the *S. aureus* bacteria better than that of *E. coli* bacteria. But overall it is being seen that the knitted fabric is slightly better than woven fabric in terms of antibacterial property. It is assumed that this happens due to the looping structure of knitted fabric.

Table 1: Evaluation of Anti-Bacterial activity in Pad-Dry-Cure Method

SL No.	Method of Coating	Fabric Type	Bacteria	Area of Fabric (mm ²)	Resisted area of			Net resisted area (mm ²)
					untreated sample (mm ²)	treated sample (mm ²)	treated sample after 3 times wash (mm ²)	
1	Pad-dry-cure	Woven	<i>S. aureus</i>	352	0	475	408	123
2		Woven	<i>E. coli</i>	352	0	408	368	56
3		Single Jersey	<i>S. aureus</i>	352	0	567	520	215
4		Single Jersey	<i>E. coli</i>	352	0	475	414	123
5		Rib	<i>S. aureus</i>	352	0	720	616	368
6		Rib	<i>E. coli</i>	352	0	450	408	98

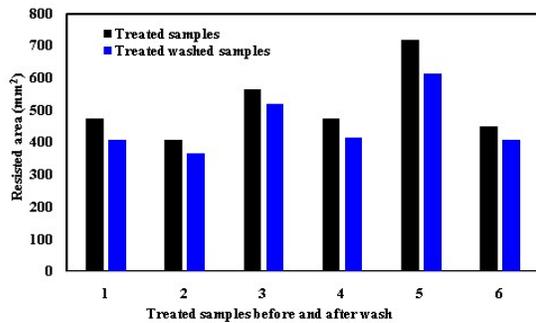


Figure 2: Antibacterial resistance of different fabrics using pad-dry-cure method.

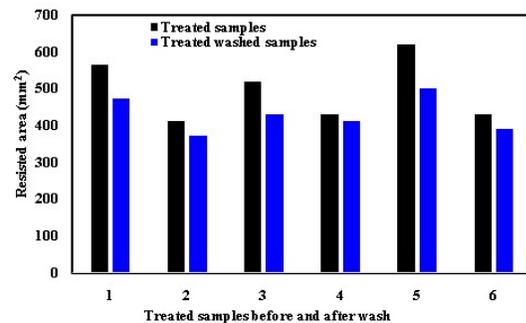


Figure 3: Antibacterial resistance of different fabrics using spin coater

Likewise Table 1, the results of three types of fabric treated with spin coating method after applying *S. aureus* and *E. coli* bacteria are presented in the table 2. The area of the sample fabric was 352 sq. mm for each. In the result it is seen that the treated fabric without wash showed the best anti-biotic properties. And there was a significant reduction in anti-microbial property after three time washing and the non-treated fabric exhibits no resistance at all.

Table 2: Evaluation of Anti-Bacterial activity in spin Coating Method

SL No.	Method of Coating	Fabric Type	Bacteria	Area of Fabric (mm ²)	Resisted area of			Net resisted area (mm ²)
					untreated sample (mm ²)	treated sample (mm ²)	treated sample after 3 times wash (mm ²)	
1	Spin	Woven	<i>S. aureus</i>	352	0	567	475	215
2		Woven	<i>E. coli</i>	352	0	414	374	62
3		Single Jersey	<i>S. aureus</i>	352	0	520	432	168
4		Single Jersey	<i>E. coli</i>	352	0	432	414	80
5		Rib	<i>S. aureus</i>	352	0	621	500	269
6		Rib	<i>E. coli</i>	352	0	432	391	80

The comparison of the three types of fabric treated with spin coating method after applying *S. aureus* and *E. coli* bacteria are presented in the figure 3. From the chart, it is clearly understood that the double jersey/rib fabric showed the maximum anti-bacterial property providing a resisted area of 621 sq. mm. And the anti-bacterial treatment resists the *S. aureus* bacteria better than that of *E. coli* bacteria. Again, it is being seen that the knitted fabric is slightly better than woven fabric in terms of antibacterial property.

However, rib/double jersey fabric treated in pad-dry-method gave 720 sq. mm area which is much higher than spin coating method. In pad-dry-cure method, the sample is immersed into solution which may provide more

fixations of chemicals into the samples than spin coating method. Hence, pad-dry-cure method may have better performance than spin coating.

4. CONCLUSIONS

From this study, it concludes that double Jersey fabric showed highest protection against bacteria than single jersey and woven fabric. Moreover, pad-dry-cure method gave good antibacterial protection than spin finish. There is more scope to test others structures of fabric like silk, polyester etc. for finding out the best antibacterial coated cloth. Although, spin coating takes less time and saves chemical solution uptake than pad-dry-cure. But setting up a spin coater for antibacterial finish is a costlier matter than pad-dry-cure method. Moreover, antibacterial activity of ZnO particles were greater on gram-positive than gram-negative bacteria. Therefore, antibacterial treatment resists the *S. aureus* bacteria better than that of *E. coli* bacteria. The antimicrobial properties of these fabrics will allow additional protection to them from bio deterioration during storage, transportation and consumption of goods and provide new opportunities to use in medical application.

REFERENCES

- Alagarasi, A., 2011. Introduction to Nanomaterials in Nanomaterials, Edition 1, Volume 7.
- Gazit, E., and Mitraki A., 2007. Plenty of Room for Biology at the Bottom: An Introduction to Bio nanotechnology, London: Imperial College Press, 105.
- Getie, S., Belay A., Reddy A.R.C., and Belay, Z., 2017. Synthesis and Characterizations of Zinc Oxide Nanoparticles for Antibacterial Applications, Journal of Nanomedicine & Nanotechnology, **8**(5), 1-8.
- Gunalan, S., Sivaraj R., and Rajendran V., 2013. Green Synthesized ZnO Nanoparticles against Bacterial and Fungal Pathogens, Progress in Natural Science, Materials International, **22**(6), 693-700.
- Hashim, A. A., 2012. Smart Nanoparticles Technology, Croatia: In Tech Janeza Trdine, 9.
- Hatamie, A., Khan A., Golabi M., Turner A.P.F., Beni V., Mak W.C., Sadollahkhani A., Alnoor H., Zargar B., Azam S., Nur O., and Willander M., 2015. Zinc Oxide Nanostructure-Modified Textile and Its Application to Biosensing, Photocatalysis, and as Antibacterial Material, Langmuir, **31**(39), 10913–10921.
- Hossain, M. B., Salam M. A., Yousuf M. A., and Rashed A., 2014. Characterization and Antibacterial Studies of the Mixed Ligands Complexes of Pd(ii) Ion with Phthalic Acid and Heterocyclic Amines, Journal of Engineering Science, **5**(1), 47-53.
- Islam, S., and Butola B.S., 2019. The Impact and Prospects of Green Chemistry for Textile Technology, ISBN: 978-0-08-102491-1, UK: Woodhead Publishing, 281.
- Jinhuan, J., Jiang P., and Jiye C., 2018. The Advancing of Zinc Oxide Nanoparticles for Biomedical Applications, **3**, 1-18.
- Khwaja, S.S., Azizur R., Tajuddin and Azamal H., 2018. Zinc Oxide Nanoparticles and Their Activity against Microbes, Nanoscale Research Letters, **13**(1), 1-13.
- Nouailhat, A., 2008. An Introduction to Nanoscience and Nanotechnology, United States: John Wiley & Sons, pp 4.
- Rajendran, R., Balakumar C., Ahammed, H.A.M., Jayakumar, S., Vaideki, K. and Rajesh, E.M., 2010. Use of Zinc Oxide Nano Particles for Production of Antimicrobial Textiles, International Journal of Engineering, Science and Technology, **2**(1), 202-208.
- Ruszkiewicz, J., Pinkas A., Ferrer B., Peres T., Tsatsakis A., and Aschner M., 2017. Neurotoxic Effect of Active Ingredients in Sunscreen Products, A Contemporary Review, Toxicology Reports, **4**, 245–259.
- Horikoshi, S., and Serpone N., 2013. Microwaves in Nanoparticle Synthesis: Fundamentals and Applications, 1st Edition, USA, Wiley & Sons, 1-24.
- Singh, G., Joyce E.M., Beddow J., and Mason T.J., 2012. Evaluation of Antibacterial Activity of ZnO Nano Particles Coated Sonochemically onto Textile Fabrics, The Journal of Microbiology, Biotechnology and Food Sciences, **2**(1), 106-120.
- Smijs, T.G., and Pavel S., 2011. Titanium Dioxide and Zinc Oxide Nanoparticles in Sunscreens: Focus on their Safety and Effectiveness, Nanotechnology, Science and Applications, **4**, 95–112.
- Textile News, Apparel News, RMG News, Fashion Trends, 2017. Antimicrobial Finishes for Textile Materials, available at: <https://www.textiletoday.com.bd/antimicrobial-finishes-textile-materials/> (Accessed 5 August 2019).