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# Facile Process for the Development of Antiviral Cotton Fabrics with Nano-Embossed Copper Oxide

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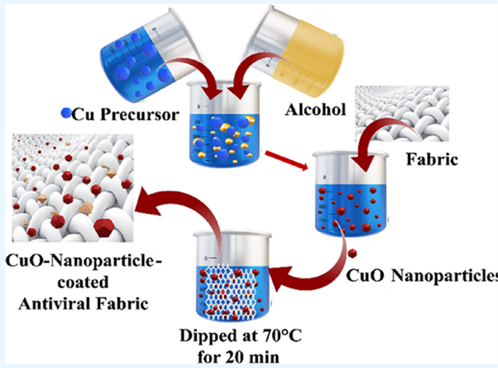
**ABSTRACT:** Metallic or metal oxide-based nanoparticles have the potential to inactivate viruses. Among various metals, copper has shown edge over others. One of the rapidly evolving areas is to combine nanoscience for production of self-sanitizing antiviral surfaces. In this study, we designed antiviral-coated fabrics to combat the spread of viruses. Copper oxide nanoparticles were sonochemically synthesized and subsequently deposited using the dip-coat process to modify the surface of fabric. The morphology and structure of uncoated and coated fabrics were examined by scanning electron microscopy, X-ray diffraction, FTIR, and elemental analysis. The findings show that small, agglomerated rugby ball structures made of copper oxide (CuO) nanoparticles ($16 \pm 1.6$ nm, according to the Scherrer equation) develop on the surface of fabric, resulting in nano-embossing and a hydrophobic (contact angle > 140°) surface. The CuO-coated fabric yielded the maximum zone of inhibition for antibacterial activity. The virucidal activity (against human adenovirus-B) of CuO nanoparticle-fabricated fabric against adenovirus shows decreased 99.99% according to the ISO 18184 testing standard. With the dip and dry approach, any textile industry can use the simple coating procedure without having to change its textile operations. This fabric can be widely used in the face mask, clothing, bedding, and aprons, and the coating remains efficient over more than 25 washes.




## 1. INTRODUCTION

Textile plays an important role in protecting humans from the worst seasonal conditions. It can also prevent the human body from the impact of thrust, sharp objects, and electric shocks.[1,2] Wearing stuff is the first thing that came to mind whenever we think about the protection of the body. However, due to the large surface area of fabric, superior ability to absorb moisture, and inclusion of fat, starch, and protein, these textiles might actually enhance microbial development rather than providing protection against microbial infection and transmission. These microbes involve everything from bacteria to viruses, including the ongoing coronavirus. Numerous scientists have applied a variety of antibacterial chemicals to textile, including synthetic and natural materials to make the textile stuff safe against microbes.[3,4] Normal textile fabric does not exhibit any antiviral properties, but if specific antiviral chemicals are integrated into the fabrics, textile can become a good protection against microbes. There are a variety of ways to include sustainable antiviral materials into fabrics, including the use of certain chemicals to reduce the surface energy of textiles[5−7] or application of antiviral compounds to the surface of the fabric. In general, surface treatment is considered good, which can be applied to a variety of fabric types, such as wool, silk, cotton, and other combinations of fabrics. These procedures include pad-dry-cure, exhaust method, microencapsulation, and these coating techniques alone or in combination.[8,9] Fabrics are frequently treated using the exhaust method of dyeing, disperse dyeing process, or pad-dry-cure to attach antiviral materials. The surface coated by using these methods can inhibit the virus by oxidizing and dissolving the lipid layer of and breaking down the virus into its fragments. Another technique is microencapsulation, which is the process of encapsulating an active chemical as the core substance inside of a polymeric shell. For antiviral coating, a microcapsule solution is made by combining antiviral and polymeric coating in an emulsion reactor for 6−48 h while being stirred and mixed at 1k to 10k rpm. In an agitator, this coating and binder are finally applied to fabrics. This method is cumbersome and is frequently used to finish medical and athletic apparel with an antibacterial finish. Another important coating technique is the deposition of antiviral material onto the surface of textile to increase surface properties, which are spray, gravure coating, roll, and rod and are most commoly used for non-woven substrates.[8,10−12] In addition, polymeric coating deposition of

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nanoparticles (organic and inorganic) on the fabric surface is evolving. A variety of methods, such as dip-coat, pad-dry-cure, and in situ synthesis of NPs using sonication and sputter-coating, are reported to deposit nanoparticles on textile substrates.[13] The deposition method and type of nanoparticle have great impact on the texturing of the fabric surface and, therefore, on the properties of surface-modified fabric. More than 30 heavy metals have great potential as biocidal, including gold (Au), silver (Ag), bismuth (Bi), copper (Cu), cobalt (Co), mercury (Hg), iron (Fe), manganese (Mn), platinum (Pt), zinc (Zn), lead (Pb), titanium (Ti), nickel (Ni), antimony (Sb), and tin (Sn).[14−20] These metals showed biocidal effects even at low concentrations. Metals like copper (Cu) and silver (Ag) are known as good antimicrobial agents for centuries.[21−32] Nowadays, a new class of nanometallic antibiotics has revolutionized the antimicrobial properties.[33−35] Several researchers have reported that Cu shows high antimicrobial efficacy and faster results when compared to Au.[36] Another metal oxide that is more biocompatible in nature and is normally used as a semiconductor in most antimicrobial applications is $TiO_2$. It is used for disinfection of viruses, bacteria, and fungi.[37,38] Thus, different metals can be used to make antiviral-coated fabrics.[6,39−42] As mentioned earlier, copper has shown remarkable properties as compared to other materials, so researchers have attempted to develop scalable processes for coating fabric with copper oxide.[43] One of the facile approach to synthesizing metal oxides without calcination in single pot is to produce metal oxide by sonicating metal acetates under basic pH.[44] By using this approach, metallic oxide nanoparticles can directly be produced by sonicating metal acetate-impregnated fabric. However, it requires specific design of equipment, which makes it difficult to be adopted in normal industrial setups. In the present study, sonochemically synthesized CuO nanoparticles are coated by the industrially compatible dip-coat method, which is as simple as a regular dyeing method. The fabric functionalized with sonochemically obtained CuO nanoparticles was characterized through different techniques including FTIR, SEM, EDS, and XRD to investigate the morphological and structural properties of synthesized material. Antiviral as well as antibacterial tests were performed to investigate the biocidal properties of material, and to check the reusability, washing test was also performed.

## 2. MATERIALS AND METHODS

All chemical reagents (up to 98.99% pure) used for the synthesis and fabrication of nanoparticles were purchased from Sigma-Aldrich, Germany. All chemicals purchased were analytical grade pure and were used without any further purification. The Gram-negative bacterial strain *Escherichia coli* (*E. coli*) (ATCC 25922, Gram-negative bacterium) was investigated for the antibacterial activity of CuO nanoparticle-fabricated fabric purchased from Sigma-Aldrich, Germany. Cotton fabric was purchased from Al-Karam, Pakistan.

**2.1. Synthesis of Copper Oxide Nanoparticles.** Copper oxide NPs were obtained by modifying a reported sonochemical method.[43] Typically, 0.1 mol of copper(II) acetate $[Cu(CO_2CH_3)_2]$ was dissolved in water and ethanol (3:7). The pH of the solution was adjusted between 8 and 9 with the help of aqueous ammonia. The solution was ultrasonicated for 90 min at the temperature of 60 °C. The color of solution turned to brown from blue, which indicated the successful synthesis of CuO nanoparticles.

**2.2. Pre-Treatment of Cotton Fabrics.** The cotton fabric ready for dyeing was selected for experimental purpose and purchased from Alkaram Studio, Pakistan. The fabric came with information by the company, which was as follows: thread counts of 600−700, 143 gsm with high breathability, medium heat retention ability, medium stretchability, and high moisture wicking ability. The fabric composition was organic fibers from cotton seeds. For functionalization of fabric with CuO nanoparticles, 6 × 6 cm pieces of cotton fabric were selected, washed with deionized water to remove any impurities, dried at room temperature, and weighed before functionalization.

**2.3. Fabrication of CuO Nanoparticles onto the Cotton Fabric.** After the successful synthesis of nanoparticles, the fabric was functionalized by using a simple "dip and dry" method. A piece of 6 × 6 cm cotton fabric was dipped into the prepared CuO nanoparticle solution. Initially, the percent uptake of CuO nanoparticles was tested by the change in UV absorption of solution (before and after uptake) and it was found that the fabric used in current study can uptake 2.89% w/w CuO. The ratio of solution to fabric was adjusted to uptake 0.90% CuO by weight into cotton fabric. The temperature of the reaction bath was adjusted at 60 °C; after 20 min, the color of fabric turned from white to brown, which indicates the successful functionalization of CuO nanoparticles into cotton fabric. The fabric was dried at room temperature and washed several times with distilled water to remove any unreactive residues. To check the stability of metal oxide nanoparticle coating, a standard washing procedure test was performed with detergent and deionized water (Figure 1).


**Figure 1.** (A) Non-coated cotton fabric and (B) cotton fabric coated with CuO nanoparticles.

**2.4. Antibacterial Activity Measurement of the CuO-Coated Fabric.** The Gram-negative bacterial strain *E. coli* (ATCC 25922, Gram-negative bacterium) was investigated for the antibacterial activity of the CuO nanoparticle-coated fabric. For this study, the disc diffusion method was used against Gram-negative *E. coli*. *E. coli* was cultured on Mueller-Hinton agar at 37 °C, and freshly cultured inoculations were collected into tubes containing 50 mL of nutrient broth for the evaluation of the effect of CuO nanoparticle-fabricated fabric on the bacterial growth curve by the disc diffusion method.[45] A 100 $\mu$L aliquot of *E. coli* was spread on the solidified surface of Mueller-Hinton agar in a Petri plate, and 6 mm CuO nanoparticle-coated fabric was placed in this Petri plate and incubated at 37 ± 1 °C for 24 h. After that, the inhibition zone was measured in mm.

**2.5. Antiviral Assays of the CuO-Coated Fabric.** The antiviral activity of the CuO nanoparticle-coated fabric was tested according to the international standard for the determination of virucidal activity of textile ISO 18184:2019.

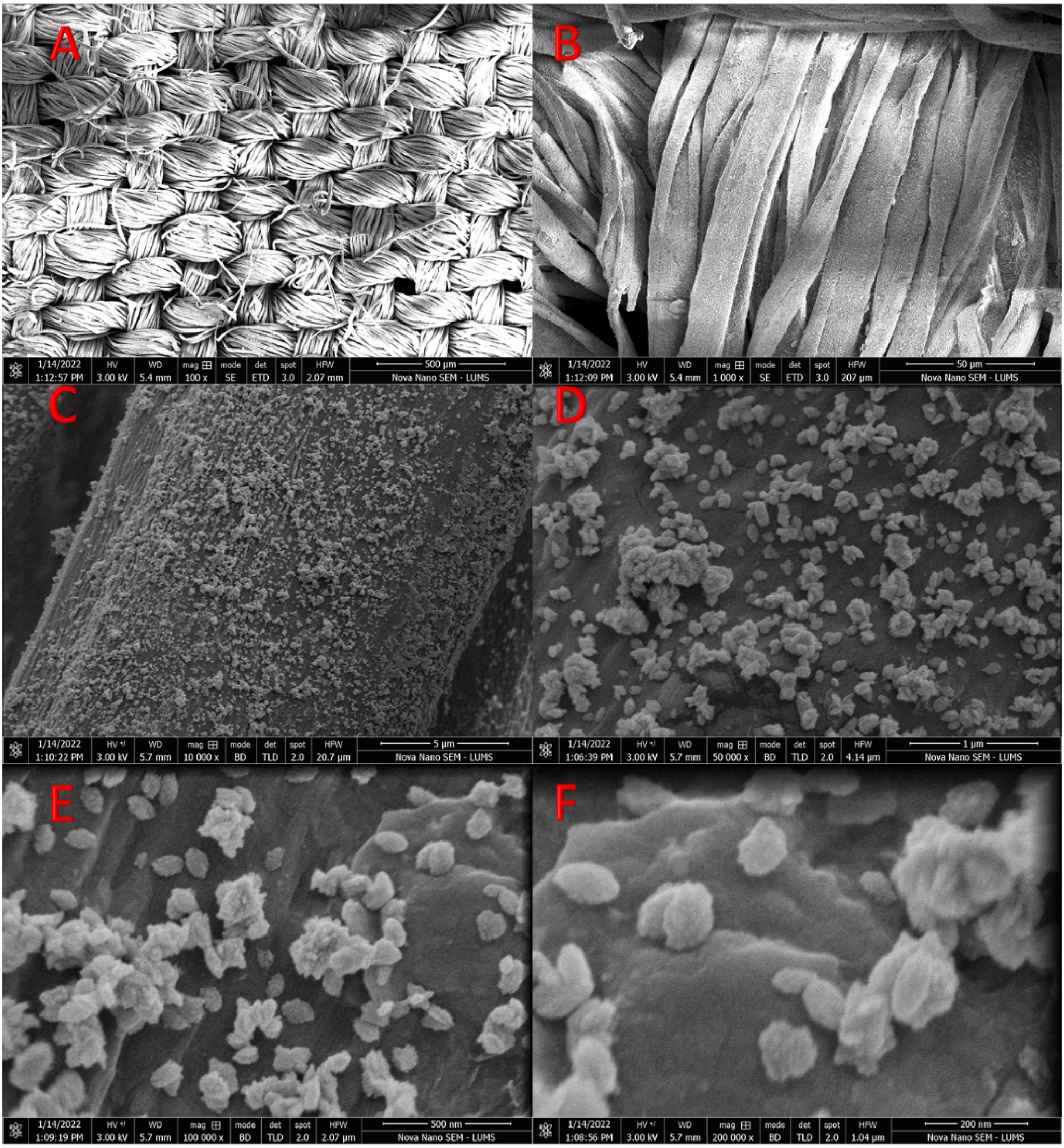


**Figure 2.** FE-SEM images of fabric coated with CuO nanoparticles at different magnifications. (A) 500 $\mu$m, (B) 50 $\mu$m, (C) 5 $\mu$m, (D) 1 $\mu$m, (E) 500 nm, and (F) 200 nm.

The method was based on the measurement of concentration of $10^5$ PFU (plaque-forming unit) of adenovirus HAdV-B. A total of 0.2 mL of adenovirus suspension was dropped on the sample of each fabric, and samples were sealed in separate glass vials. Samples were kept for 2−18 h at 25 °C; after the required time, 20 mL of SCDLP was added to each vial and the vial was well shaken by using a vortex mixer. After mixing, 1.8 mL of Eagle's minimal essential medium (EMEM) was added to 0.2 mL of suspension, which was transferred for plaque essay.

## 3. CHARACTERIZATION OF THE CUO-COATED FABRIC

The synthesized fabric coated with metal oxide nanoparticles was characterized with different analytical techniques. The X-ray diffraction analysis was performed by using a Shimadzu XD-3A XRD. The scanning rate was fixed at a step of 0.6 s within the range of 5 to 79 2$\theta$ value by using graphite-monochromatized Cu as the anode at 25 °C with a k-alpha/k-beta ratio of 0.5. For the identification of crystallinity of



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material, the XRD data was analyzed by using X'Pert High Score software. Morphological characteristics were analyzed through FE-SEM. A Nova NanoSEM 450 Field-Emission Scanning Electron Microscope was used with resolution in high vacuum mode with a 3 kV TLD and ETD detector with SE mode. FTIR spectroscopy was used to study the functional group of synthesized fabric at room temperature equipped with a diamond crystal. The spectra were recorded between the wavenumbers of 400 and 4000 $cm^{-1}$. The model of the instrument used was a Thermo Scientific Nicolet TM iS10. The morphological characteristics of synthesized fabric were studied by using a scanning electron microscope, and elemental composition was obtained by energy dispersive X-ray analysis (EDX) from a Hitachi S2300 operated at 2 to 20 kV. Sonication was performed with Elma, $E_{30H}$, Elmasonic, Germany, having a stainless-steel bath with an operating frequency of 37 kHz and an ultrasound power of 80 W.

## 4. RESULTS AND DISCUSSION

**4.1. SEM and EDX Analyses.** The morphological studies for the CuO-coated fabric were carried out using SEM analysis (Figure 2), which shows that the nanoparticles are mixed, irregular, semi-spherical-shaped, agglomerated (60−100 nm in size), and distributed throughout the threads of fabric. These semispherical nanoparticles are lined up as a structure like porous globular nano-rugby balls individually or arranged in various agglomerates of individual nano-rugby balls. The shape of nanoparticles is found different in this study compared to the sonochemically synthesized CuO nanoparticles, which looked like flakes.[46] This may be due to difference in the deposition process used in this study. In the reported study, CuO nanoparticles were synthesized in situ on fabric, whereas in this work, already synthesized nanoparticles were deposited by the dip-coating method. The elemental composition of fabric coated with CuO nanoparticles was investigated through EDX analysis. The percentages of C, O, and Cu were found to be 48.12, 45.07, and 13.17%, respectively. The signal of Au is due to the sample coated with gold. EDX confirmed that CuO nanoparticles were successfully fabricated into the fabric (Table 1).

**Table 1. Percentages of Elements Present in the Fabric Coated with CuO Nanoparticles**

| element | line type | apparent concentration | k ratio | weight % |
|---|---|---|---|---|
| C | K series | 17.2 | 0.17198 | 40.12 |
| O | K series | 43.26 | 0.14557 | 45.07 |
| Cu | L series | 5.91 | 0.05913 | 13.17 |
| total | | | | 100 |

**4.2. FTIR Analysis.** The CuO-coated fabric was characterized by FTIR analysis for surface functionalities (Figure 3). The characteristic peaks were observed at 590 ,670, 1120, 1451, 1639, 2890, and 3265 $cm^{-1}$. The peaks present between 400 and 700 $cm^{-1}$ are correlated to metal oxides. The sharp peak observed at 590 $cm^{-1}$ is the characteristic peak of bond formation of Cu−O. Similarly, the peak at 1120 $cm^{-1}$ is assigned to bending/stretching, which is present in the Cu−O nanoparticles.[47] The peak detected around 3200−3500 $cm^{-1}$ may be attributed to the stretching vibration of OH or NH groups.[48] The broad absorption band at 3265 $cm^{-1}$ reveals the presence of water molecules or amine from ammonia solution used to prepare nanoparticles; the same solution was used to dip-coat the fabric. Likewise, the peaks around 1600−1670 $cm^{-1}$ represent the bending vibration of N−H bonds. The peak at 1639 $cm^{-1}$ indicates the presence of the N−H group on the cotton fabric. The peak at 2890 $cm^{-1}$ corresponds to the stretching of the single bond of C−H assigned to the alkyl group. The FTIR study confirms the presence of amine, alkane, alcohol group, and water molecules on the fabric material as well as formation of a metallic nanostructure on the surface.

**4.3. XRD Analysis.** Figure 4 shows the PXRD peaks of CuO (coated fabric). A doublet was observed at 15° value, followed by a peak with the highest peak intensity at 22.5°, which corresponds to cotton fabric.[49] The peaks at 35.5 ± 0.2° and 38.0 ± 0.2° are the characteristic peaks of the monoclinic crystal of CuO. The PXRD pattern obtained here is similar to as-prepared CuO nanoparticles obtained using the sonochemical process by Wongpisutpaisan et al.[50]

The particle size of the CuO nanoparticles was estimated with the help of Scherrer equation (eq 1)

$$D = \frac{0.9\lambda}{\beta \cos \theta} \quad (1)$$

where $D$ is the size of particles, 0.9 is the constant, $\lambda$ is the X-ray wavelength, $\theta$ is the Braggs angle, and $\beta$ is the width half maximum. The size of synthesized CuO nanoparticles was found to be 16 ± 1.6 nm by using the Scherrer equation. The calculated size of crystallinity of material from XRD data does not match the sizes deduced from the SEM images, because SEM shows the agglomerated particle size, not the crystallite size. Therefore, it is inferred that agglomerates are comprised of four or five CuO crystallites.

**4.4. Antibacterial Activity of the CuO-Coated Fabric.** The antibacterial activity of the CuO nanoparticle-coated fabric was carried out against *E. coli*. Ten times washed fabric was used to evaluate the efficiency. Figure 5 shows the zone of inhibition for *E. coli*; no colonies were seen where the fabric is placed, with a zone of inhibition of 3.6 mm (Table 2), which is double that of the positive control (streptomycin).

**4.5. Antiviral Activity of the CuO-Coated Fabric.** The antiviral activity of the CuO nanoparticle-coated fabric was carried out against adenovirus (HAdV-B) by using ISO 18184:2019.[51] Ten times washed fabric was used to evaluate the efficiency to understand the activity. The treated fabric is cut into 20 × 20 mm in size. These pieces of antiviral fabric were inoculated in 200 μL of adenovirus (HAdV-B) and left at 25 °C for 2 h. Two milliliters of washout solution is added in each vial and agitated for five times for 5 s to wash out virus. After washing out from specimens and incubation time, $TCID_{50}$ was calculated (eq 2) with comparison to control fabric according to the equation given below. The results are shown in Table 2, which shows an antiviral activity of 99.99% against adenovirus in 2 h.

$$M = \lg(V_b) - \lg(V_c) \quad (2)$$

where $M$ is the antiviral activity, $\lg(V_b)$ is the logarithm average (three infectivity titers with the reference specimen (PFU/vial) for 2 h), and $\lg(V_c)$ is the logarithm average (three infectivity titers contacting with the antiviral fabric for 2 h).

Percentage reduction was calculated (eq 3) as follows,

$$\text{reduction } (\%) = ((V_b - V_c)/V_b) \times 100 \quad (3)$$

where $V_b$ = PFU/vial (average of three infectivity titers while contacting with reference fabric for specific time) and $V_c$ =


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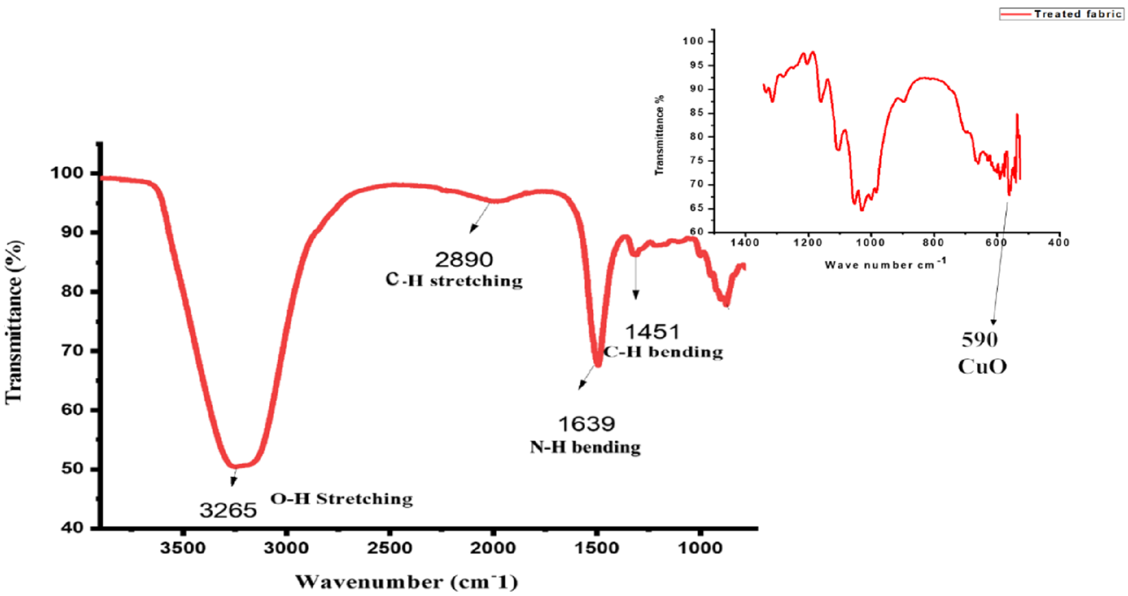


Figure 3. FTIR spectra of the synthesized CuO-fabricated fabric.

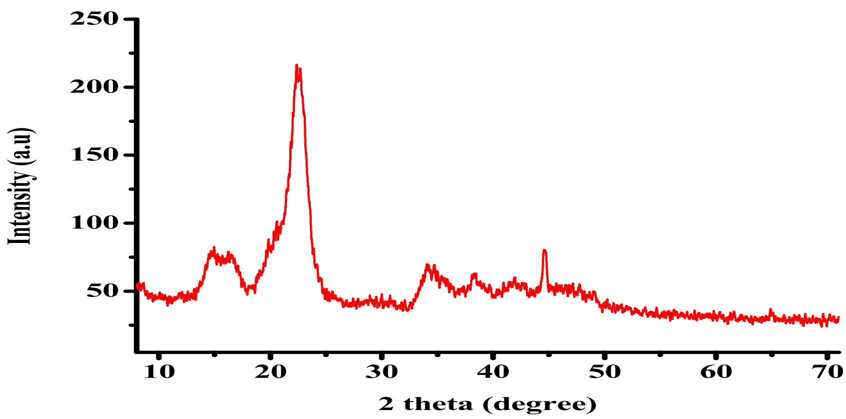


Figure 4. PXRD spectrum of the CuO NP-coated fabric.

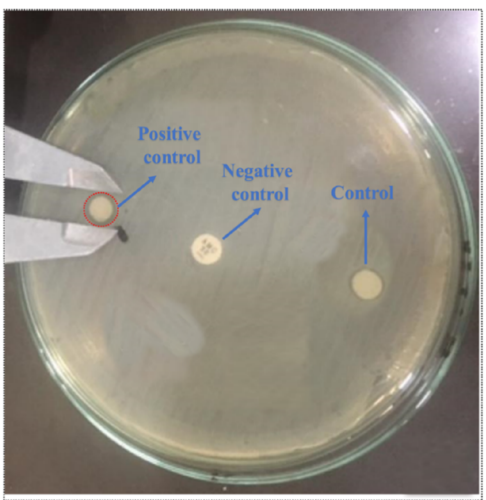


Figure 5. Zone of inhibition in diameter (mm). The sample showed an inhibition zone against *Escherichia coli* (*E. coli*), which is significantly larger against 10 $\mu$L of positive control antibiotic streptomycin.

Table 2. Antiviral and Antibacterial Activity Results for the CuO-Coated Fabric

| virus/microbe type | strain | test method | reference standard | results |
|---|---|---|---|---|
| bacteria | *Escherichia coli* (*E. coli*, ATCC 1555) | AATCC 100-1999 | streptomycin | zone of inhibition, 6.6 mm |
| virus | human adenovirus (HAdV-A12) | ISO-18184 | HAdV-B | 99.99% |

PFU/vial (average of three infectivity titers contacting with antiviral fabric for specific time).

**4.6. Wash Durability Test.** The durability of antiviral properties of fabric coated with CuO nanoparticles can be evaluated through wash cycles. Figure 6 shows the wash cycle and weight % decrease during washing. The wash cycle was conducted by initially washing fabric with simple distilled water for 15 min at room temperature and drying.[52] For wash cycle calculations, the AATCC 61 (2A) international method was used. Stainless steel beakers were used, which were filled with 150 mL of detergent solution and 50 stainless balls. These beakers were placed inside a laundry machine for 45 min at 40



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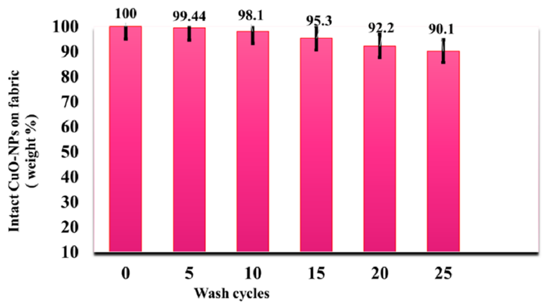


**Figure 6.** Wash cycle durability of CuO nanoparticle fabric.

rpm and 50 °C. After the washing cycle was complete, the fabric was rinsed with distilled water and dried at room temperature.[53] The results show the slight decrease in the number of CuO nanoparticles from fabric due to the release of CuO nanoparticles, but the fabric has retained its antiviral as well as antibacterial properties up to 25 washes. The wash cycle stability indicates that the particles are tightly bound to the surface of fabric. The CuO particles may be bound on the surface of fabric by interlinking on OH groups of cellulose. The alkaline medium helps create OH on the surface of cotton fabric,[54,55] and addition of ammonia can form various reactive functionalities as shown below.[56]

$$C_6H_7O_2(OH)_3 + NH_4OH(aq) \rightarrow C_6H_7O_2(OH)_3NH_4OH$$

CuO particles seem to be attached through ammonium linkage as also suggested by the FTIR peak at 1639 $cm^{-1}$, which indicates the presence of N−H functionality, or by the OH link by forming N−Cu and O−Cu bonds, which impart durability to the CuO coating on cotton fabric.

**4.7. Antiviral Mechanism of the CuO-Coated Fabric.** Nanoparticles show excellent antiviral activity toward many strains of viruses because of their unique properties. The mechanism of action of a metal ion or metallic nanoparticles can be the attachment or connection of nanoparticles to the surface of virus, making the viral cell unable to be attached to the host cell; production of toxic highly reactive oxygen species that could attach to the spike or membrane of virus and inhibit the function of nucleic acid and protein; enhancement of the immune system of the host by stimulating the nucleus; and destruction of host cell and inhibition of the spread of virus.

Copper acts differently on different types of viruses. Therefore, the prepared CuO-coated fabric was investigated for various properties like the possibility of producing free radicals and the hydrophobicity of the surface. Free radical generation was assessed using the methylene blue method, and hydrophobicity was evaluated by measuring the contact angle. Methylene blue solution (100 mg/L, 100 mL) was contacted with a 7 $cm^2$ piece of CuO-coated fabric for 5 h. No change in absorption was observed in methylene blue as compared to the uncoated fabric, which suggests that the CuO-coated fabric does not generate free radicals (Figure 7). However, the reduction in absorbance as compared to pure methylene blue dye is due to adsorption of dye on fabric itself.

Results for contact angle measurements are shown in Figure 8. The ability of a solid surface to be wetted by a liquid can be classified by its wetting contact angle according to the contact angle: the surface whose contact angle is <90° is hydrophilic, those with a contact angle between 90° and 150° are hydrophobic, and those surfaces with a contact angle of >150° are called super-hydrophobic. The contact angle was calculated by using ImageJ software. The result shows that the contact angle of treated fabric was found to be 142°; hence, the fabric is characterized as hydrophobic.

It is well established that an increase in the hydrophobicity of the surfaces including fabrics would generally improve their antimicrobial activity. Hydrophobic surfaces can better be able to interact with viruses and destabilize the microbial membrane. It may be suggested that the fabric is hydrophobic and nano-embossed (SEM image, Figure 2) with tiny nano-rugby balls of CuO-agglomerated nanoparticles. Limited study on the mechanism of action of this fabric toward adenovirus-B shows that the surface roughness of fabric with CuO creates reactive interference with the capsid proteins of adenovirus. Electrostatic attraction between Cu and NH/SH may be suggested as a possible route for inactivation of adenovirus.

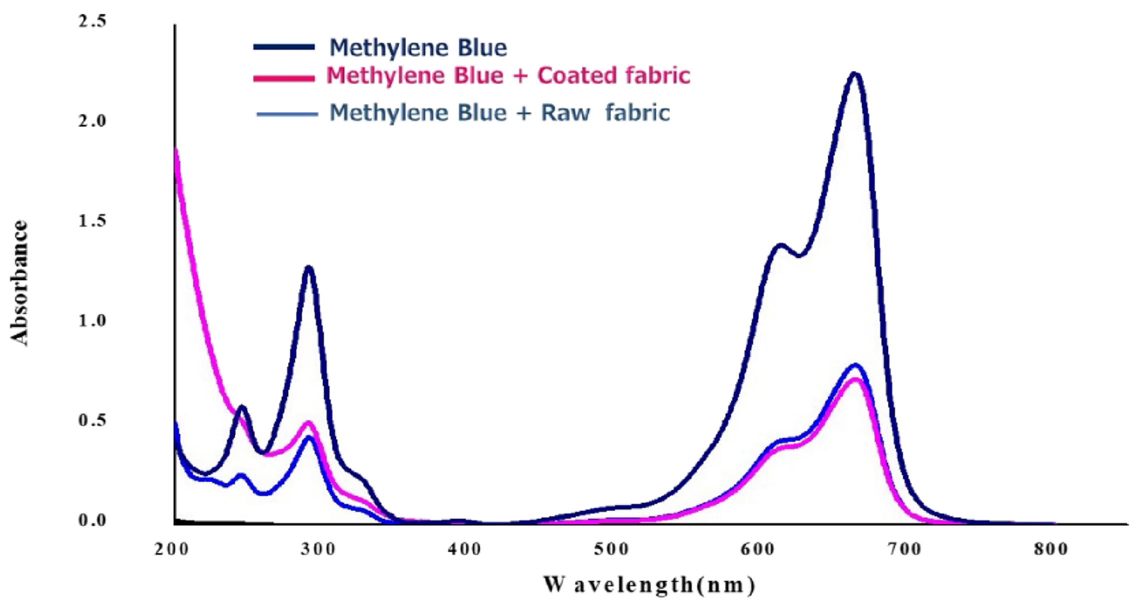


**Figure 7.** UV−Vis spectra of methylene blue in solution and in contact with treated and untreated fabrics.


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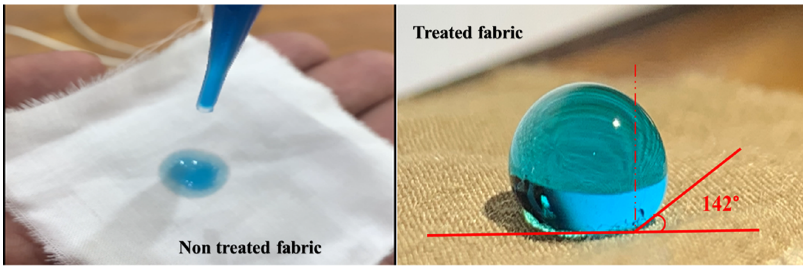


**Figure 8.** Contact angle measurement of non-treated and treated fabrics.

## 5. CONCLUSIONS

According to the findings, sonication is a successful way for creating copper oxide nanoparticles in a single step, which are then strongly bonded to cotton fabric using an approach that is suitable for use in industry (dip and dry method). The CuO nanoparticles remained in the nanometer range of size even after deposition and formed a uniform layer and continuous surface coverage on cotton fabric. The treated fabric developed hydrophobic characteristics due to the nanotexturing effect by CuO nanoparticles. Both the virucidal and antibacterial activities of the CuO nanoparticle-coated fabric against adenovirus and Gram-negative bacteria exhibit encouraging results. Furthermore, copper is cheap and readily available, which is therefore industrially viable for production of antiviral fabrics using the process reported in this research. If employed in surface modification of fabric in the form of masks, aprons, lab coats, bedsheets, and casual apparel, this antiviral fabric can play a significant role in rendering the virus inactive.

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### Notes

The authors declare no competing financial interest.

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