gels

MDPI

*Article*

# The Highly Durable Antibacterial Gel-like Coatings for Textiles

**Seyedali Mirmohammadsadeghi [1], David Juhas [2], Mikhail Parker [1], Kristina Peranidze [1], Dwight Austin Van Horn [3], Aayushi Sharma [4], Dhruvi Patel [4], Tatyana A. Sysoeva [4], Vladislav Klepov [5] and Vladimir Reukov [1,*]**

1 Textiles, Merchandising and Interiors, University of Georgia, Athens, GA 30605, USA
2 Independent Researcher, Webster, NY 14580, USA
3 Independent Researcher, Summerville, SC 29486, USA; avanhorn@chitozanhealth.com
4 Department of Biological Sciences, University of Alabama in Huntsville, Huntsville, AL 35758, USA; tatyana.sysoeva@uah.edu (T.A.S.)
5 Department of Chemistry, University of Georgia, Athens, GA 30605, USA; klepov@uga.edu
* Correspondence: reukov@uga.edu

**Abstract:** Hospital-acquired infections are considered a priority for public health systems since they pose a significant burden for society. High-touch surfaces of healthcare centers, including textiles, provide a suitable environment for pathogenic bacteria to grow, necessitating incorporating effective antibacterial agents into textiles. This paper introduces a highly durable antibacterial gel-like solution, Silver Shell™ finish, which contains chitosan-bound silver chloride microparticles. The study investigates the coating's environmental impact, health risks, and durability during repeated washing. The structure of the Silver Shell™ finish was studied using transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDX). The TEM images showed a core–shell structure, with chitosan forming a protective shell around groupings of silver microparticles. The field-emission scanning electron microscopy (FESEM) demonstrated the uniform deposition of Silver Shell™ on the surfaces of the fabrics. AATCC Test Method 100 was employed to quantitatively analyze the antibacterial properties of the fabrics coated with silver microparticles. Two types of bacteria, *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*), were used in this study. The antibacterial results showed that after 75 wash cycles, a 100% reduction for both *S. aureus* and *E. coli* in the coated samples using crosslinking agents was observed. The coated samples without a crosslinking agent exhibited 99.88% and 99.81% reductions for *S. aureus* and *E. coli* after 50 washing cycles. To compare the antibacterial properties toward non-pathogenic and pathogenic strains of the same species, MG1655 model E. coli strain (ATCC 29213) and a multidrug-resistant clinical isolate were used. The results showed the antibacterial efficiency of the Silver Shell™ solution (up to 99.99% reduction) coated on cotton fabric. AATCC-147 was performed to investigate the coated samples' leaching properties and the crosslinking agent's effects against *S. aureus* and *E. coli*. All coated samples demonstrated remarkable antibacterial efficacy, even after 75 wash cycles. The crosslinking agent facilitated durable attachment between the silver microparticles and cotton substrate, minimizing the release of particles from the fabrics. Color measurements were conducted to assess the color differences resulting from the coating process. The results indicated fixation values of 44%, 32%, and 28% following 25, 50, and 75 washing cycles, respectively.

**Keywords:** antibacterial coatings; gels; biomedical textiles; chitosan; silver microparticles



**Citation:** Mirmohammadsadeghi, S.; Juhas, D.; Parker, M.; Peranidze, K.; Van Horn, D.A.; Sharma, A.; Patel, D.; Sysoeva, T.A.; Klepov, V.; Reukov, V. The Highly Durable Antibacterial Gel-like Coatings for Textiles. *Gels* **2024**, *10*, 398. https://doi.org/10.3390/gels10060398

Academic Editor: Andrea Zille

Received: 10 May 2024
Revised: 6 June 2024
Accepted: 7 June 2024
Published: 13 June 2024



## 1. Introduction

Infectious diseases continue to be one of the leading causes of death around the world and are a priority in global public health [1,2]. Based on a recent study, infection-related deaths were the second leading cause of death, accounting for 13.7 million mortalities in 2019. To clarify more, between 10 and 18% of all main causes of death were attributed

to 33 bacterial pathogens, including the resistant strains, which can mainly be found in contaminated areas, e.g., clinical wards [3].

Hospital-acquired infections (HAIs), as a fourth primary cause of death in the United States, have imposed a considerable burden on healthcare systems in both human resources and economic issues [4]. High-touch surfaces in hospitals and other health centers, such as floors, beds, tables, and textile-based materials, are at risk of being contaminated by pathogenic bacteria. They can easily transfer infectious microbes to patients and hospital workers and cause additional problems [5]. Drug-resistant infections spreading via textile surfaces pose serious threats, and eliminating bacteria from such surfaces has the potential to drastically reduce the spread. To tackle this issue of contaminated hospital textile materials, the common ways are to use disinfectants to remove surface germs and kill bacteria or use disposable items several times a day. Both methods are burdensome for healthcare systems since they are time-consuming and less effective than expected. The high transmission rate of pathogens can still infect patients and healthcare workers with weakened immune systems. In addition, antibiotic overuse is increasing, which can cause the emergence of resistant bacteria and various environmental pollutions [6]. Therefore, antibacterial surfaces, particularly in textiles, are becoming increasingly desirable to protect against different types of microorganisms [5,7–15].

Various hospital textiles, such as privacy drapes, upholstery, lab coats, and scrub suits, can be an ideal environment for a broad range of microorganisms, such as bacteria and fungi, to live for months [7]. These textiles are prone to being contaminated by bacteria and transfer them to the human body [8]. The porosity of these materials creates good conditions for microorganisms to thrive by providing moisture and warmth [5,8,9].

There are different types of antibacterial agents that could be used to create antimicrobial textiles. These include chemical compounds, natural extracts, and metal nanoparticles. Chlorinated aromatic structures, N-halamines, and Quaternary Ammonium compounds are the most common chemical compounds for antibacterial fabric finishing in the textile industry [16].

As a chlorinated aromatic structure, one of the chemical antimicrobial agents, Triclosan, has been used in different products, such as toothpaste, soaps, shampoos, and facial cleaners. Based on the hydrophobic nature of Triclosan, it has been incorporated into various plastic-based materials, including textiles and air filters. It alters the integrity of the microbe's membranes and disrupts bacterial fatty acid synthesis by inhibiting the activity of enzymes, which are involved in its synthesis. Triclosan's antibacterial properties are affected by its concentration and the coating process of treated materials. Although it showed higher antibacterial properties when it was concentrated in some studies, the bacteria utilize the multidrug-resistance (MDR) efflux pump to expel the drug, thereby reducing its activity. The efficacy of the efflux pump decreased at lower concentrations, and it did not allow the bacteria to form colonies. However, the main drawbacks of using Triclosan are related to environmental concerns. The presence of Triclosan in the environment can accumulate in aquatic organisms, such as fish and algae. It disrupts the endocrine system of aquatic organisms, affecting their growth and reproduction and increasing bacteria resistance against Triclosan [17,18].

N-halamines, such as 2,2,5,5-Tetramethyl-Imidozalidin-4-One (TMIO), create covalent bonds with nitrogen and halogen, offering broad-spectrum antibacterial activity [19,20]. Another amine-based N-halamine, bis(N-chloro-2,2,6,6-tetramethyl-4-piperidinyl) sebacate (Cl-BTMP), was used as the antimicrobial agent due to its potent antimicrobial efficiency, along with good light and thermal stabilities [21]. Quaternary Ammonium compounds (QACs) disrupt microbial cell membranes through positively charged nitrogen, exerting biocidal effects against various microorganisms [22,23]. While these compounds offer effective antibacterial properties, concerns arise regarding their environmental impact, potential health risks, and durability during washing or exposure to high temperatures. The selection of antibacterial compounds for fabric finishing requires a careful balance between efficacy, environmental considerations, and durability [16,24].

Natural substances, such as plant extracts, essential oils, and animal products, provide various antimicrobial properties. The investigations of the antimicrobial properties of various natural dyes extracted from plants have increased the variety of antimicrobial agents, contributing significantly to the production of antibacterial fabric. Natural plant extracts, such as curcumin, basil, clove oil, cyclic oligosaccharides, and sericin, and natural plants, including onion, aloe vera, and pomegranate, have shown unique antimicrobial properties [25–27]. Caffeic acid and quercetin as two types of material extracted from plants have also demonstrated antibacterial properties [28,29]. Natural plant extracts offer potential benefits, but they have some considerable drawbacks. One key limitation is the variability in composition and potency, influenced by factors like plant growth conditions and extraction methods, leading to an inconsistent efficacy. The spectrum of the antimicrobial activity may be limited, and the concentrations for effectiveness can vary. Degradation over time affects stability and shelf life, while the solubility, bioavailability, and formulation challenges may impede practical application.

Using metals and metal salts, such as silver, copper, gold, zinc oxide, and titanium dioxide, for their antimicrobial properties in textiles has a rich historical background. They can be toxic to pathogens, even at a very low concentration. Titanium dioxide and zinc oxide are employed for their bactericidal properties, with $TiO_2$ acting as a photocatalyst that requires UV radiation for antibacterial applications. ZnO offers a cost-effective agent with superior whitening and UV-blocking properties on textiles. However, challenges such as the need for UV radiation and potential limitations in killing microbes exist for these coating agents. Copper has excellent antibacterial properties and poses a biocidal effect. While it is not as effective as silver for antibacterial action, copper is still considered a promising material to be incorporated into different substrates, including textiles [16,30,31].

Nanoparticles, particularly silver nanoparticles (AgNPs), are prominent for their broad-spectrum antimicrobial activity, self-cleaning properties, and increased dyeability. AgNPs have demonstrated effectiveness against various microbes. Its antibacterial effectiveness is superb compared with other metal-based materials, such as Cu-based ones, ZnO, and $TiO_2$. The minimum bactericidal concentration for silver is lower than other metal-based antibacterial agents, including copper-based ones. It can even have an antibacterial efficiency in a small amount [30]. In contrast to antibiotics, which use specific mechanisms of action against pathogens, the biocidal effect of silver nanoparticles has been reported with different mechanisms. One mechanism of action is that due to the electrostatic attraction of silver ions to sulfur proteins, they can adhere to the cell wall and cytoplasmic membrane of bacteria, which increases the permeability of the cytoplasmic membrane and disrupts the bacterial envelope [7,8,32,33].

The challenges associated with the production and coating processes of AgNPs include agglomeration, achieving the desired morphology and the production of a uniform particle size. Agglomeration decreases the stability and efficacy of AgNPs, which needs effective reagents to overcome this challenge [2]. The common reducing agents, such as sodium dodecyl sulfate and ascorbate, are toxic and hazardous and cause serious environmental problems. Capping with natural compounds has emerged as a solution to promote the stability of AgNPs, particularly when applied to textiles. However, weak interactions on the fabric surface are considered a significant drawback. To address this, the impregnation of capping agents, whether synthetic polymers or natural biological agents, becomes important for enhancing the stability of AgNPs on textiles. Among the various capping agents, chitosan, as a biodegradable and biocompatible biopolymer, also has weak antibacterial and antifungal properties [34–36]. A range of mechanisms were reported that mainly involves penetrating the phospholipid bilayer of the bacterial membranes and disrupting the cytoplasmic membrane [37]. Chitosan can also be an ideal reagent, which mitigates agglomeration by introducing electrostatic repulsion and steric hindrance between AgNPs [1]. Moreover, due to having active functional groups in the chitosan structure, it forms hydrogen bonds with cotton, hydrolyzed polyester, and nylon fabrics [38]. This multi-functionality of chitosan makes it a promising candidate for achieving stable,

well-dispersed, and antimicrobial AgNP-coated textiles, which can tackle the challenges in both the production and long-term stability fields [39]. The application of chitosan as a coating agent or stabilizer for silver nanoparticles has been reported in several studies that demonstrated better stability and antimicrobial efficacy [2,10,37,40–43]. For instance, Siva et al. synthesized chitosan–silver nanocomposites with an average particle size of 10 nm to study their antibacterial activity against *Staphylococcus aureus* and *Escherichia coli* [37].

A non-leaching functional textile is generally preferred because it retains its antimicrobial activity for an extended period. Moreover, releasing significant amounts of antimicrobial agents into the environment can be an environmental issue [16]. Therefore, in this study, a gel-like solution containing chitosan-bound silver chloride microparticles (AgMPs) was developed in two distinct average sizes to investigate their impacts on antibacterial properties, considering concerns about their environmental impact when they are nano-sized. Our focus included the durability and effectiveness of cotton fabrics coated with silver–chitosan over multiple washes. *Staphylococcus aureus*, *Escherichia coli*, and multidrug-resistant strains (MG and ES) were chosen to study the efficiency of the coated fabrics.

## 2. Results and Discussion

### *2.1. Silver Shell™ Solution Characterization*

The sample of Silver Shell™ solution was annealed at 80 °C over 2 h to obtain a film for the phase composition study. The PXRD data collected for the material are demonstrated in Figure 1. The diffractogram illustrates the presence of AgCl, $Ag_2O$, and Ag phases, which corresponds to the method of solution synthesis developed at Chitozan Health LLC. The synthesis includes the precipitation of AgCl submicron particles into a chitosan solution that stabilizes the particles by increasing the zeta potential and reduces silver ion migration from silver particles. Narrow characteristic peaks corresponding to the identified phases indicate the regions of high crystallinity.

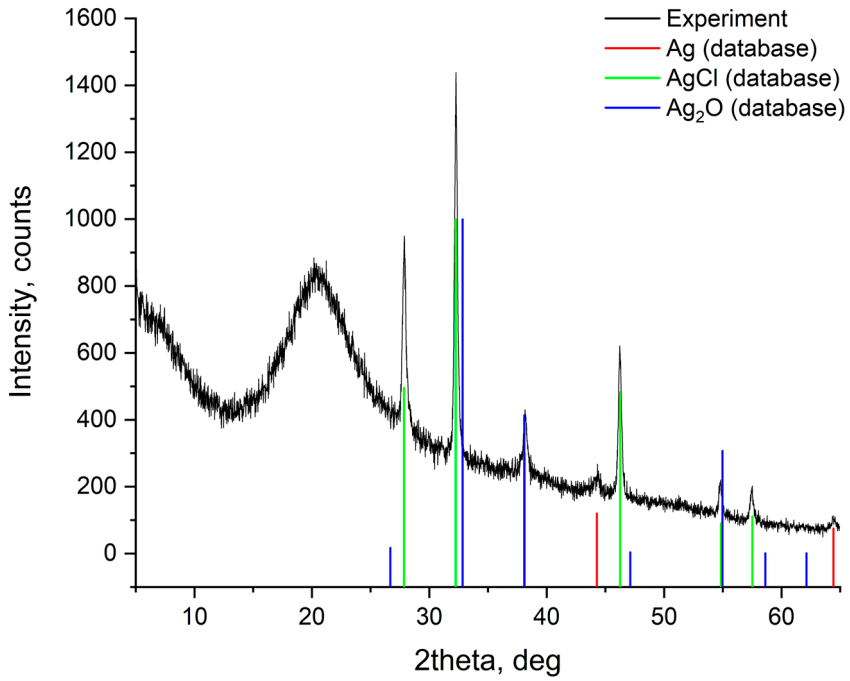


**Figure 1.** PXRD plot for the annealed Silver Shell™ solution.

The TEM images of synthesized SS26 and AgMP034 are shown in Figure 2a,b, respectively. According to Figure 2a, the submicron particles were scattered separately and primarily exhibited a spherical morphology. However, some multi-shaped particles were also observed, which might have been the result of aggregation during the preparation of particles for TEM analysis. The TEM image of AgMP034 (Figure 2b) showed a core–shell

structure, in which chitosan formed a protective shell around groupings of microparticles. Based on the TEM images, the median size of the particles was found to be 100 nm for SS26 (Figure 2c) and around 202 nm for AgMP034 (Figure 2d). The formation of a chitosan shell is advantageous due to its ability to form hydrogen bonds with the functional groups present in materials like cotton or nylon, thereby facilitating specific interactions and enhancing the coating stability.

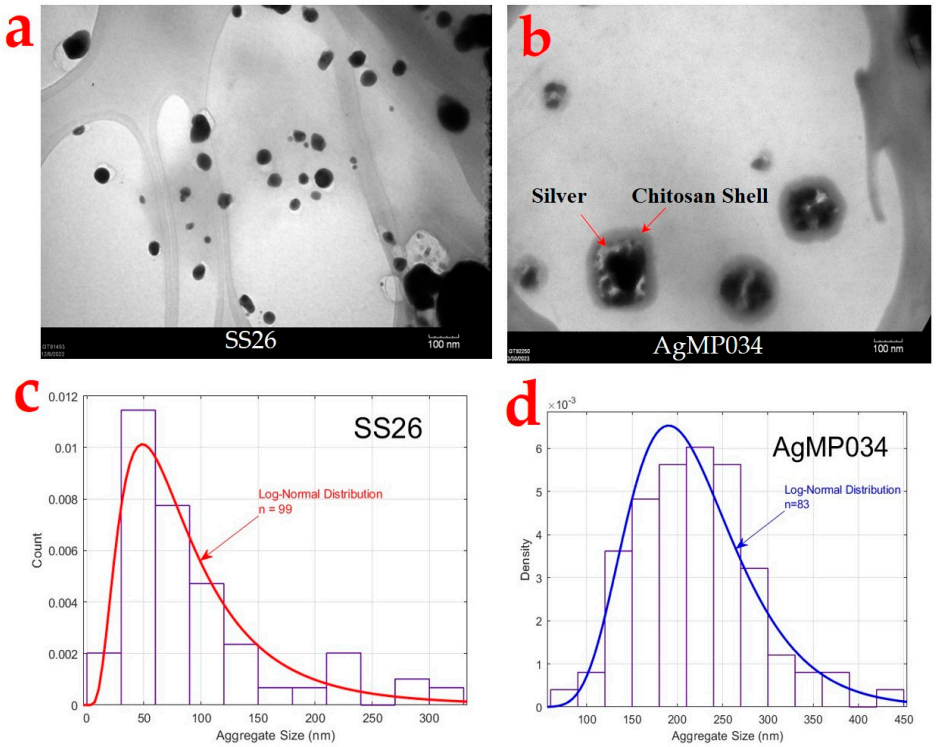


**Figure 2.** TEM images of (**a**) SS26 and (**b**) AgMP034; log-normal distributions of (**c**) SS26 (n = 99) and (**d**) AgMP034 (n = 83).

The analysis of the EDX (Figure 3) showed the presence of Ag and Cl elements, which were related to the precipitation of AgCl.

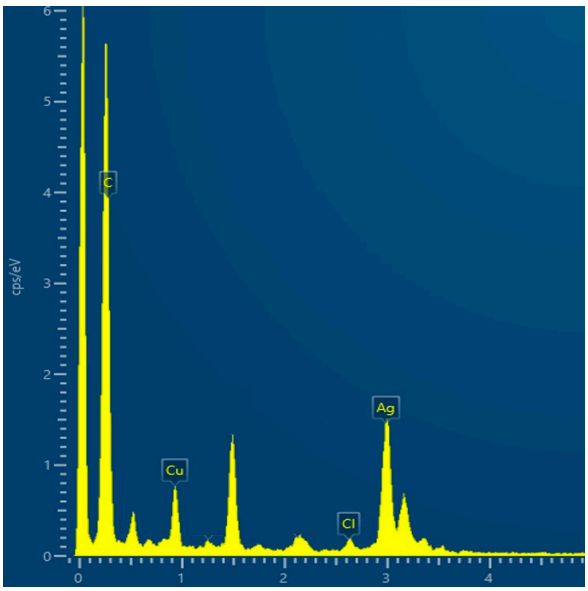


**Figure 3.** EDX analysis of the synthesized Silver Shell™ solution on a copper grid.

### 2.2. Coating Characterization

Cotton fabrics coated with Silver Shell™ solution were chosen in this work. Generally, cotton and silver particles display physical adsorption [9]. The morphological changes in cotton fabrics coated with Silver Shell™ after 25, 50, and 75 washing cycles were monitored by FE-SEM. As shown in Figures 4a and 5a, the Silver Shell™ was uniformly deposited on the cotton fabric surface. The surface of the cotton fabric was relatively smooth after the coating. After washing the samples, Figures 4b–d and 5b–d show that the Silver Shell™ began to detach, along with the cotton fibers, with the most noticeable detachment observed after 75 washing cycles. However, for SS26, there were some detachments after 50 washing cycles. The results showed that the Silver Shell™ had a strong bond with cotton fabrics due to the presence of hydrogen bonding between the chitosan and cotton.

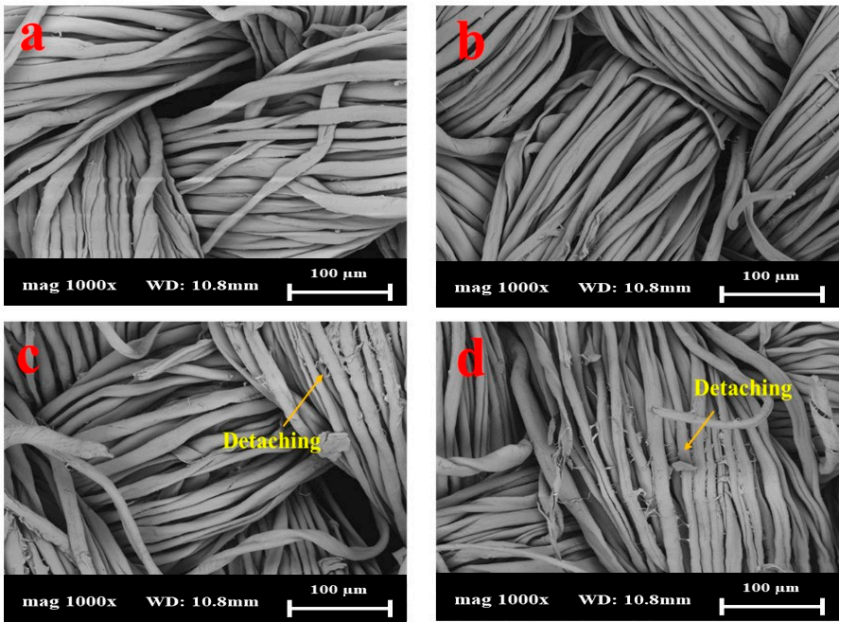


**Figure 4.** SEM images of SS26 with different washing cycles: (**a**) no washing, (**b**) 25 cycles, (**c**) 50 cycles, and (**d**) 75 cycles.

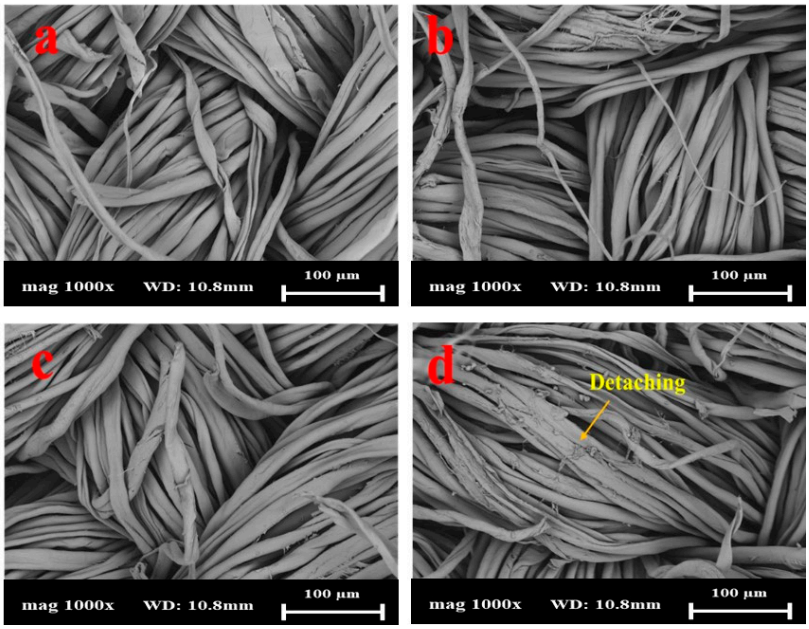


**Figure 5.** SEM images of AgMP034 with different washing cycles: (**a**) no washing, (**b**) 25 cycles, (**c**) 50 cycles, and (**d**) 75 cycles.

Figure 6 shows the result of the EDX analysis of AgMP034 without washing. The weight percentage of Ag was 0.2%, as the silver coverage on coated fabric was very low.

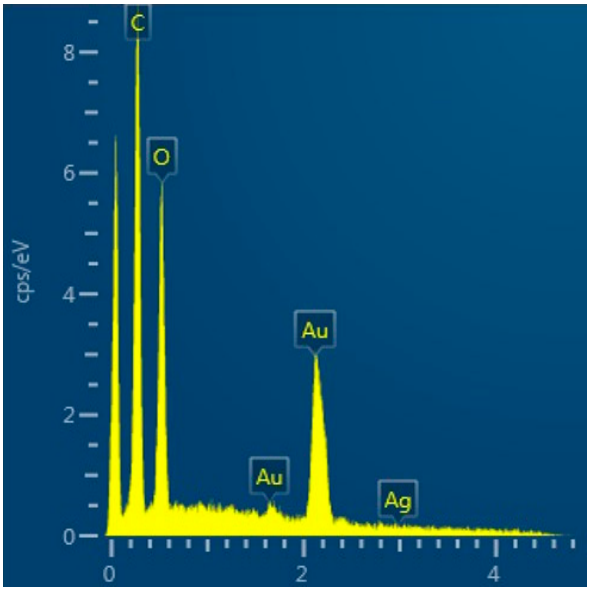


**Figure 6.** EDX analysis of AgMP034 without washing.

### *2.3. Antibacterial Studies*

Cotton fabrics, as one of the most common types of textiles, are prone to the growth of microorganisms. Dangerous pathogens, particularly bacteria, can flourish in these fabrics and cause several problems [44]. Therefore, antibacterial textiles are essential in highly contaminated areas, including hospitals, as they can minimize the spread of pathogens. The main goal of this study was to develop highly durable textile fabrics with antimicrobial properties, even after multiple washes. This was achieved by coating the cotton fabrics with a gel-like silver–chitosan solution. To investigate the antibacterial properties of the coated fabric, AATCC 100 (quantitative analysis) and AATCC 147 (qualitative analysis) were utilized. Table 1 shows the killing efficiencies of AgMP034 and SS26 after undergoing 75 wash cycles, and also A34 without washing and after 25 and 50 cycles following the AATCC 100 method. Untreated cotton fabric was also used as a control sample. According to Table 2, all treated samples, except A34, after 50 washing cycles showed a 100 percent reduction in both *E. coli* and *S. aureus*, even after 75 wash cycles. Despite the decreased silver content due to washing, the remaining quantity surpassed the minimum Ag concentration necessary to sustain their antibacterial properties. Our preliminary studies indicate that 100% bacterial reduction on coated cotton fabrics required a minimum Ag coverage of around 7.5 μg/g of fabric.

**Table 1.** AATCC100—effects of crosslinking and washing cycles on antibacterial properties.

| Samples | Washing Cycles | *E. coli* | | *S. aureus* | |
|---|---|---|---|---|---|
| | | Percent Reduction | Log 10 Reduction | Percent Reduction | Log 10 Reduction |
| **Control** | 0 | 0 | 0 | 0 | 0 |
| **A34** | 0 | 100 | >6 | 100 | >6 |
| | 25 | 100 | >6 | 100 | >6 |
| | 50 | 99.88 | 2.927 | 99.81 | 2.723 |
| **AgMP034** | 0 | 100 | >6 | 100 | >6 |
| | 25 | 100 | >6 | 100 | >6 |
| | 50 | 100 | >6 | 100 | >6 |
| | 75 | 100 | >6 | 100 | >6 |

**Table 1.** *Cont.*

| Samples | Washing Cycles | *E. coli* | | *S. aureus* | |
|---|---|---|---|---|---|
| | | Percent Reduction | Log 10 Reduction | Percent Reduction | Log 10 Reduction |
| SS26 | 0 | 100 | >6 | 100 | >6 |
| | 25 | 100 | >6 | 100 | >6 |
| | 50 | 100 | >6 | 100 | >6 |
| | 75 | 100 | >6 | 100 | >6 |

**Table 2.** AATCC100—effects of coating and washing cycles on multidrug-resistant *E. coli* strains.

| Samples | Washing Cycles | *E. coli MG1655* | | *ESBL* | |
|---|---|---|---|---|---|
| | | Percent Reduction | Log 10 Reduction | Percent Reduction | Log 10 Reduction |
| Control | 0 | 0 | 0 | 0 | 0 |
| A34 | 0 | 99.971 | >3 | 99.99 | >4 |
| | 25 | 99.976 | >3 | 99.99 | >4 |

To investigate the effect of the crosslinking agent on the durability of coated fabrics, sample A34 was prepared. Table 1 showed that after 50 wash cycles, the reduction percent decreased to 99.88 (log 10 reduction of 2.927) and 99.81 (log 10 reduction of 2.723) for *E. coli* and *S. aureus*, respectively. The lack of the crosslinking agent caused a weaker interaction between the silver particles and cotton fabrics.

The antibacterial properties of the uncoated and silver-coated cotton fabrics were also analyzed through the AATCC 147 method. Parallel streaks of both *S. aureus* as Gram-positive and *E. coli* as Gram-negative bacteria were drawn in separate Petri dishes and subsequently covered with the pieces of the samples. Figure 7 shows the photographs of the negative control and control inoculated with bacteria as a reference. The bacterial growth that occurred underneath the uncoated fabrics was visible. Also, there was no bacterial growth on the agar for the negative control, indicating that the incubator's environment and samples were clean. Based on Figure 8, sample A34 without washing and after 25 wash cycles showed no growth of bacteria, which indicated splendid antibacterial activities. However, after 50 wash cycles, there were bacteria colonies, which may be attributed to the lack of enough silver coverage on this sample. In fact, due to the lack of crosslinking agent, silver particles coated with chitosan lost their stability on the fabric and were washed away during laundering. For sample A34 without washing, a slight inhibition zone was observed around the fabric, indicating the leaching of silver particles. This likely resulted from a weak interaction between the coating material and fabric, leading to a gradual release by washing. However, there was no distinct boundary for sample A34 after 25 wash cycles.

To study the effect of the crosslinking agent on the washing durability, the antibacterial activities of AgMP034 and SS26 were also qualitatively tested against the same bacterial strains used for previous samples, and the results are shown in Figure 9 for *E. coli* and Figure 10 for *S. aureus*. Based on Figures 9 and 10, all the coated samples showed remarkable antibacterial efficacy, even after 75 wash cycles, which can be attributed to the sufficient Ag coverage on the samples. Therefore, highly durable coated fabrics can be obtained by using a crosslinking agent. The crosslinking agent helped to form a stable and durable coating on the cotton fabrics. It improved the adherence of the microparticles to the cotton fibers, resulting in sufficient antibacterial properties, even after several washing cycles.

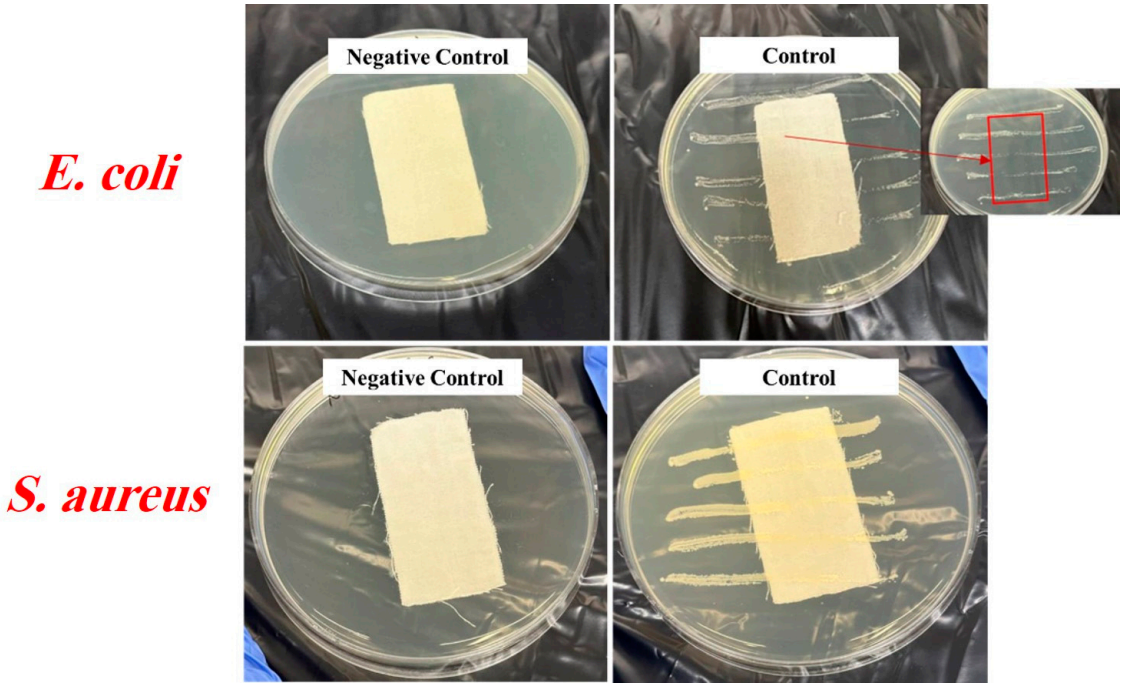


**Figure 7.** AATCC 147 for negative control and untreated fabric against *E. coli* and *S. aureus*.

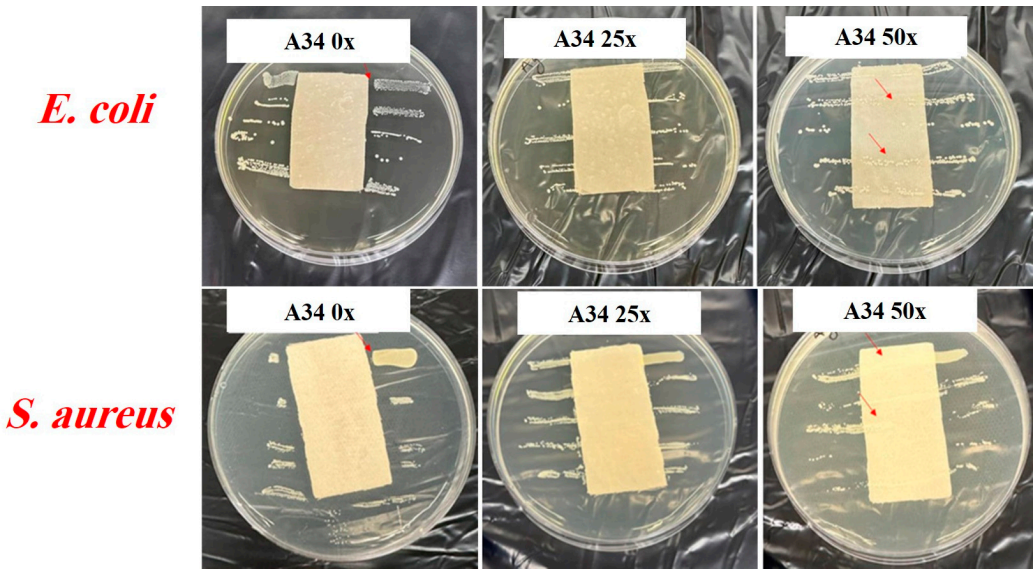


**Figure 8.** AATCC 147—A34 sample with no washing and 25 and 50 wash cycles against *E. coli* and *S. aureus*. Red arrows show the growth of bacteria.

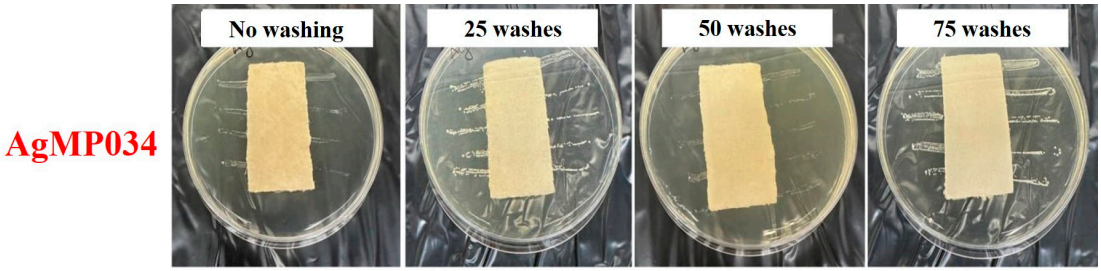


**Figure 9.** *Cont.*

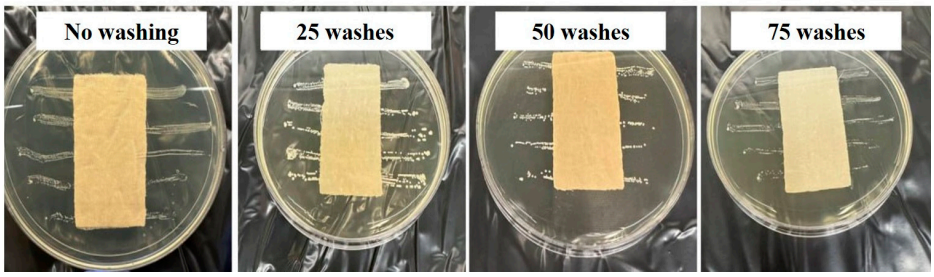


**Figure 9.** AATCC 147—AgMP034 and SS26 samples with no washing and 25, 50, and 75 wash cycles against *E. coli*.

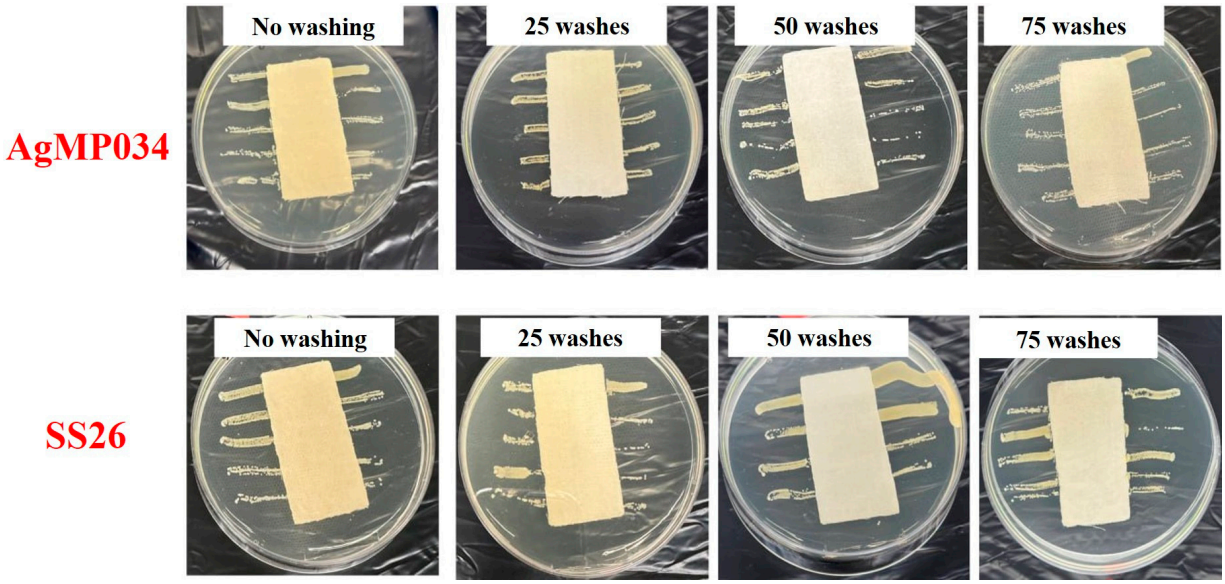


**Figure 10.** AATCC 147—AgMP034 and SS26 samples with no washing and 25, 50, and 75 wash cycles against *S. aureus*.

### *2.4. Bactericidal Action against Multidrug-Resistant E. coli Strains*

We conducted pilot experiments that compared the non-pathogenic *E. coli* MG1655 strain and the extended-spectrum b-lactamase-producing ESBL41 and ESBL146 *E. coli* isolates. Samples of unbleached muslin coated with the Silver Shell™ (A34) were tested according to the AATCC 100 protocol described above. According to Table 2, we found a strong (over 3 log) reduction for all tested strains regardless of using freshly coated textile samples or those that were washed 25 times. This confirmed prior tests with model *E. coli* and *S. aureus* strains (Table 1) and showed that in this setup, the Ag bactericidal action was similar toward antibiotic-sensitive and antibiotic-resistant ESBL isolates.

### *2.5. Color Measurement Study*

The color measurements were carried out to study how the color of the cotton fabrics was impacted by coating with gel-like Silver Shell™ solutions. These values are defined as L* for lightness, a* for red (+) to green (−), and b* for yellow (+) to blue (−) [9]. Tables 3 and 4 show the CIE L*a*b* values for SS26 and AgMP034 after 75 washing cycles, respectively. The color coordinates were also measured for the untreated cotton for comparison. Based on the results, by applying the fabrics with Silver Shell™, L* was decreased and both a* and b* were increased. Although there was a slight change for a*, the significant change was attributed to b*, which showed higher values for treated samples. Therefore, the bluish appearance of untreated cotton would change to yellowness.

**Table 3.** CIE L*a*b* values of SS26 samples.

| Samples | L* | a* | b* | ΔE |
|---|---|---|---|---|
| Untreated cotton | 95.73 ± 0.10 | 1.01 ± 0.19 | −3.95 ± 1.44 | 0.52 ± 0.23 |
| SS26-0x | 79.27 ± 0.39 | 2.85 ± 0.34 | 5.21 ± 0.39 | 0.96 ± 0.38 |
| SS26-25x | 87.63 ± 0.75 | 2.56 ± 0.36 | 7.29 ± 1.14 | 1.45 ± 0.56 |
| SS26-50x | 90.24 ± 0.22 | 1.69 ± 0.05 | 5.43 ± 0.68 | 0.99 ± 0.55 |
| SS26-75x | 92.52 ± 0.26 | 0.53 ± 0.13 | 4.83 ± 0.27 | 0.59 ± 0.21 |

**Table 4.** CIE L*a*b* values of AgMP034 samples.

| Samples | L* | a* | b* | ΔE |
|---|---|---|---|---|
| Untreated cotton | 95.73 ± 0.10 | 1.01 ± 0.19 | −3.95 ± 1.44 | 0.52 ± 0.23 |
| AgMP034-0x | 82.24 ± 0.38 | 2.41 ± 0.18 | 9.97 ± 1.30 | 1.91 ± 1.01 |
| AgMP034-25x | 88.71 ± 0.20 | 1.73 ± 0.29 | 5.59 ± 1.30 | 1.51 ± 0.71 |
| AgMP034-50x | 90.54 ± 0.62 | 0.76 ± 0.05 | 5.94 ± 0.42 | 1.12 ± 0.42 |
| AgMP034-75x | 93.13 ± 0.18 | 0.24 ± 0.07 | 3.89 ± 0.29 | 0.51 ± 0.20 |

The quality of the coating was evaluated based on the CIE L*a*b* color difference formula as follows [45]:

$$\Delta E*_{ab} = \sqrt{\Delta L*^2 + \Delta a*^2 + \Delta b*^2} \quad (1)$$

Measurements were randomly taken at five different locations and the color difference between each pair of these five spots was calculated, resulting in the average values presented in Tables 3 and 4. Based on the results, the color difference between SS26 and AgMP034 was below 2. This suggests a slight color difference, indicating a uniformity in the coating. The average values of color difference for SS26 and AgMP034 before washing were 0.96 and 1.91, respectively.

Figure 11 illustrates the color strength (K/S) plots of SS26 and AgMP034 before and after washing. Based on Figure 11, the color strength decreased after washing for both samples, indicating a reduced silver amount on the fabrics. To elucidate the significance of the bond formation between microparticles and cotton fabric as a substrate, the %Fixation was compared for AgMP034 and SS26 following multiple wash cycles. According to Figure 12, SS26 exhibited %Fixation values of 44%, 32%, and 28% following 25, 50, and 75 washing cycles, respectively. For AgMP034, the %Fixation values were higher at 54%, 44%, and 32% for the same sequence of washing cycles. The presence of chitosan surrounding the silver particles might have been the reason for the higher %Fixation values for AgMP034.

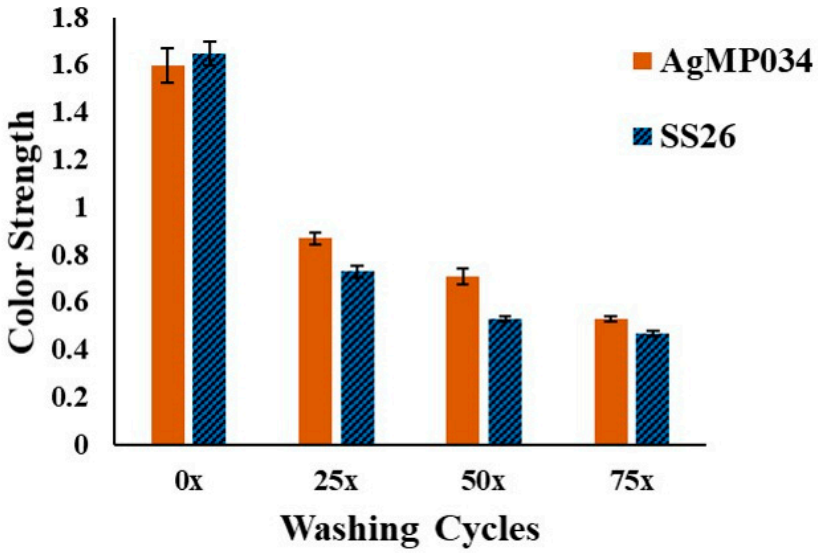


**Figure 11.** Color strengths of AgMP034 and SS26 samples in different washing conditions.

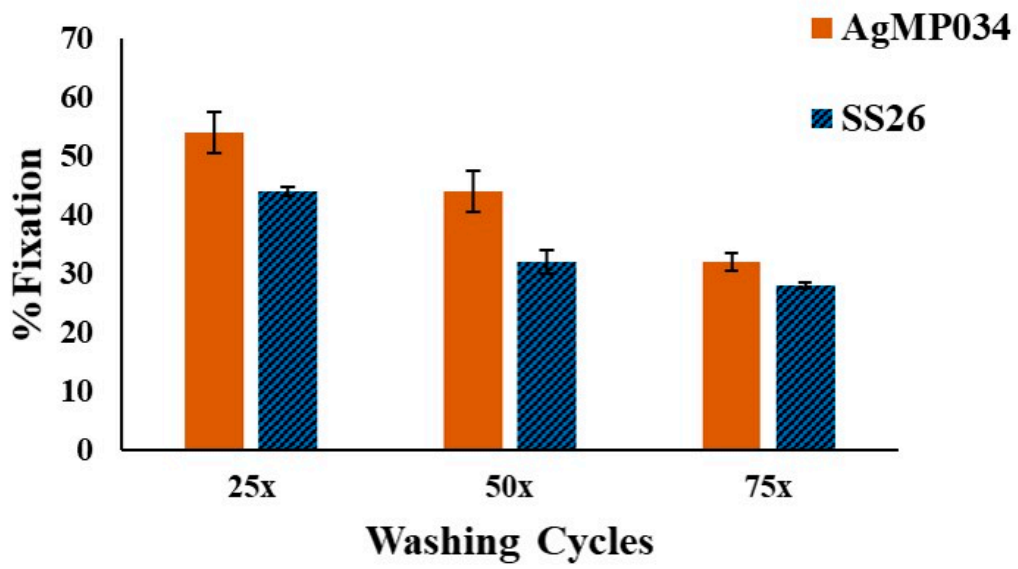


**Figure 12.** %Fixation of AgMP034 and SS26 samples in different washing conditions.

## 3. Conclusions

In summary, the present study provided data on the structures and antibacterial efficiency of coated cotton fabrics using Silver Shell$^{TM}$ solution. The results indicate the valuable and innovative Silver Shell™ solution could combat dangerous hospital-acquired infections and ensured long-term antimicrobial activity in textiles. According to the TEM results, the Silver Shell™ solution had a core–shell structure, and with chitosan, formed a protective shell around silver microparticles. The coating quality of the fabrics was investigated by FESEM before and after multiple wash cycles, indicating highly durable washing fastness. The AATCC-100 antibacterial results indicated a 100% reduction for two types of bacteria, *S. aureus* and *E. coli*, even after 75 wash cycles. The antibacterial results for the coated samples without using a crosslinking agent exhibited 99.88% and 99.81% reductions for *S. aureus* and *E. coli*, respectively, after 50 wash cycles. To investigate the leaching properties of the coated samples with Silver Shell$^{TM}$ solution and study the effect of the crosslinking agent, AATCC-147 was performed. Despite the remarkable antibacterial efficacy of coated samples, even after 75 washing cycles, the crosslinking agent minimized the release of submicron particles from the fabric. Additionally, the results of AATCC-100 on non-pathogenic E. coli MG1655 and extended-spectrum β-lactamase-producing ESBL41 and ESBL146 showed up to a 99.99% reduction in all tested strains. According to the results of color measurements, uniform coatings were obtained in different samples. Fixation values of SS26 samples showed 44%, 32%, and 28% following 25, 50, and 75 washing cycles, respectively. For AgMP034, the %Fixation values were higher than SS26 and calculated at 54%, 44%, and 32% for the same sequence of washing cycles. The presence of chitosan surrounding the silver particles might have been the reason for the higher fixation values for AgMP034.

## 4. Materials and Methods

### *4.1. Materials*

The fabrics used in this study were LS Bleached Cotton Muslin (120 GSM) from Joanne Fabrics. These fabrics were coated with two types of batches. The first was Silver Shell$^{TM}$ batch SS26 obtained from Chitozan Health LLC and produced at a facility in Chesterfield, UK. The second one was Silver Shell$^{TM}$ batch AgMP034 obtained from Chitozan Health LLC, Rochester, NY, USA. The fabrics were coated with AgMP034 and SS26, along with Glutaraldehyde (Sigma-Aldrich, St. Louis, MO, USA) as a crosslinking agent. Non-ionic surfactant NP-9 (Pfaltz & Bauer, Waterbury, CT, USA) was used for wettability improvement. To investigate the effect of the crosslinking agent, one sample, coded as A34, utilized

the same batch of AgMP034 but without adding a crosslinking agent during the coating process. The synthesis and coating process employed in this study is proprietary of the Chitozan Health LLC and utilizes silver chloride crystals suspended in a chitosan solution with adjustable sizes from 50 to 350 nm. Tryptic Soy agar was purchased from Merck, Germany. LB was purchased from Sigma-Aldrich, USA. AATCC 1993 standard reference detergent was used for washing the samples.

### *4.2. Characterization of Silver Shell™ Solutions*

The phase identification of thin-film samples containing core–shell microparticles was conducted based on powder X-ray diffraction (PXRD) data (Bruker D2 PHASER diffractometer with Cu K$\alpha$ radiation over a 2$\theta$ range of 5–65° and step size of 0.02°). A transmission electron microscope JEM100CX-II (JEOL, Ltd., Tokyo, Japan) operated at 100 kV was used to study the core–shell microparticles. Moreover, the elemental composition was analyzed by a transmission electron microscope (Hitachi-SU9000EA) equipped with energy-dispersive X-ray spectroscopy. A drop of a solution was placed on a copper grid and dried overnight at room temperature.

### *4.3. Coating Procedure*

A total of 50 g of coating solution with 7 drops of a 10% NP-9 surfactant solution were blended with 3.75 g of a 5% glutaraldehyde solution (0.375 g of glutaraldehyde per 1 g of chitosan). The swatches (20 in × 20 in) were placed into the solution and swished around to absorb as much solution as possible. After full saturation, the wet swatches were removed and wrung tightly (~3 times) to remove excess liquid. The residual liquid was removed by placing the swatches between paper towels and rolling a rubber roller over the entire surface. The samples were left at room temperature (24 °C) for 5 days to dry completely.

### *4.4. Characterization of Coated Fabrics*

#### 4.4.1. Surface Morphology

The surface morphology of the uncoated and coated fabrics was observed by field emission scanning electron microscopy (FESEM) using a Thermo Fisher Scientific (FEI) Teneo. The Teneo is equipped with energy-dispersive spectroscopy (EDX) that provides elemental analysis. The samples underwent gold coating for improved imaging using the SPI-Module Sputter coater instrument for one minute.

#### 4.4.2. Color Measurements

The color coordinates of CIE lab (L*, a*, and b*) for both untreated cotton and Silver Shell™ coated samples were conducted at 5 different places of each one using a Macbeth Color Eye 7000A Spectrophotometer. L* is related to lightness, a* is a reddish/greenish factor, and b* corresponds to the yellowness/bluish factor [46].

The color strength (K/S) of the samples coated with Silver Shell™ was also measured at all wavelengths from 400 to 700 nm (10 nm interval). This value was reported for each sample at 370 nm, which was the maximum absorbency wavelength. The color strength value was determined through the color matching software's internal computations by employing the Kubelka–Munk equation [47]:

$$\frac{K}{S} = \frac{(1-R)^2}{2R}, \quad (2)$$

where K represents absorption, S denotes scattering, and R stands for reflectance.

#### 4.4.3. Antimicrobial Fabric Test

To analyze the antibacterial properties of the silver fabric coating against uncoated controls, AATCC Test Method 100–2012 was performed. *Staphylococcus aureus* (ATCC 6538) and *E. coli* (ATCC 10536) were used as the main test organisms. Uncoated and silver-coated fabric samples were cut as circular swatches, 4.8 cm in diameter. Four swatches were

soaked with 1 mL of inoculum, containing approximately $10^6$ CFU, and placed in sterile 100 mL glass jars. Swatches soaked with 1 mL of sterile PBS were used as controls.

Jars were divided into groups: one with zero-time incubation time to quantify the initial number of viable cells, and another with overnight incubation. In the zero-time incubation group, 100 mL of sterile PBS was added immediately after placing the bacteria-inoculated swatches. After vigorous shaking, we made serial dilutions of 1/1, 1/10, 1/100, and 1/1000 from 100 µL of the PBS wash of the swatches. Then, 100 µL of each dilution was spread on agar plates in duplicates. Inoculated plates were incubated overnight at 37 °C. The colonies were counted to estimate the densities of viable cells in colony-forming units (CFU) per mL of solution (CFU/mL).

A group of jars with overnight incubation was first incubated overnight at 37 °C and then underwent the same serial dilution procedure as the zero-time group to establish the cell densities (CFU/mL).

The resulting CFU on agar plates were counted manually and the percentage of reduction (R) was calculated as follows:

$$R\% = \frac{(B - A)}{B} \times 100 \quad (3)$$

where A is the colony-forming units (CFU/mL) coated fabrics after 24 h and B is the mean of the CFU counted from control samples and coated samples at zero-time contact.

To compare the antibacterial properties toward non-pathogenic and pathogenic strains of the same species, we used the MG1655 model *E. coli* strain (ATCC 29213) and multidrug-resistant clinical isolate from the Sysoeva lab collection (extended-spectrum b-lactamase-producing ESBL41 and ESBL146 strains (Lopatkin et al., 2016)). The loading of the swatches, incubations, and plating for the CFU counting were done analogously to the procedure for other strains as described. Initially, the cell cultures of *E. coli* strains in LB were grown for 24 h, reaching an OD of 2.6–3.8 (measured by spectrophotometer Genesys 50), which is equivalent to about 2.6–3.8 × $10^8$ CFU/mL. Cells were diluted to bring them to $10^6$ CFU/mL to load $10^6$ per 5 swatches in 1 mL of solution.

To investigate the leaching of fabric silver coating against uncoated controls, we used AATCC Test Method 147–2016. Using a wire loop, test microorganisms (*S. aureus* and *E. coli*) were streaked on agar plate in 5 streaks spaced approximately 1 cm apart from each other. For each agar plate, the wire loop was submerged only once. As a result, each lower streak contained fewer CFU than the upper one. Plates were done in duplicates. After that, rectangular swatches (25 mm × 50 mm) were pressed on the streak inoculum, and the plates were incubated for 24 h at 37 °C. After incubation, the bacterial growths under and on the edges of the fabric were investigated.

**Author Contributions:** Conceptualization, S.M., D.J. and V.R.; methodology, S.M. and V.R.; software, S.M.; validation, V.R., T.A.S. and D.A.V.H.; formal analysis, S.M., D.J. and V.K.; investigation, S.M., D.J., K.P., A.S., D.P., M.P. and V.K.; resources, D.A.V.H. and V.R.; data curation, V.R. and K.P.; writing—original draft preparation, S.M.; writing—review and editing, V.R., K.P. and T.A.S.; visualization, S.M., D.J., A.S., D.P., T.A.S. and V.K.; supervision, V.R. and D.A.V.H.; project administration, V.R. and D.A.V.H.; funding acquisition, D.A.V.H. All authors have read and agreed to the published version of the manuscript.

**Funding:** Research reported in this publication was supported by the National Institute of Allergy and Infectious Diseases of the National Institutes of Health under award number R41AI172693. The content is solely the responsibility of the authors and does not necessarily represent the official views of the National Institutes of Health.

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Not applicable.

**Data Availability Statement:** All data are available within the manuscript.

**Conflicts of Interest:** The authors declare no conflicts of interest.

## References


1. Jena, P.; Mohanty, S.; Mallick, R.; Jacob, B.; Sonawane, A. Toxicity and antibacterial assessment of chitosan-coated silver nanoparticles on human pathogens and macrophage cells. *Int. J. Nanomed.* **2012**, *7*, 1805–1818. [CrossRef]
2. Canama, G.J.C.; Delco, M.C.L.; Talandron, R.A.; Tan, N.P. Synthesis of Chitosan-Silver Nanocomposite and Its Evaluation as an Antibacterial Coating for Mobile Phone Glass Protectors. *ACS Omega* **2023**, *8*, 17699–17711. [CrossRef]
3. Ikuta, K.S.; Swetschinski, L.R.; Aguilar, G.R.; Sharara, F.; Mestrovic, T.; Gray, A.P.; Weaver, N.D.; E Wool, E.; Han, C.; Hayoon, A.G.; et al. Global mortality associated with 33 bacterial pathogens in 2019: A systematic analysis for the Global Burden of Disease Study 2019. *Lancet* **2022**, *400*, 2221–2248. [CrossRef]
4. Hicks, A.L.; Reed, R.B.; Theis, T.L.; Hanigan, D.; Huling, H.; Zaikova, T.; Hutchison, J.E.; Miller, J. Environmental impacts of reusable nanoscale silver-coated hospital gowns compared to single-use, disposable gowns. *Environ. Sci. Nano* **2016**, *3*, 1124–1132. [CrossRef]
5. Gonçalves, R.A.; Ku, J.W.K.; Zhang, H.; Salim, T.; Oo, G.; Zinn, A.A.; Boothroyd, C.; Tang, R.M.Y.; Gan, C.L.; Gan, Y.-H.; et al. Copper-Nanoparticle-Coated Fabrics for Rapid and Sustained Antibacterial Activity Applications. *ACS Appl. Nano Mater.* **2022**, *5*, 12876–12886. [CrossRef]
6. Qiu, H.; Si, Z.; Luo, Y.; Feng, P.; Wu, X.; Hou, W.; Zhu, Y.; Chan-Park, M.B.; Xu, L.; Huang, D. The mechanisms and the applications of antibacterial polymers in surface modification on medical devices. *Front. Bioeng. Biotechnol.* **2020**, *8*, 910. [CrossRef]
7. Balakumaran, M.D.; Ramachandran, R.; Jagadeeswari, S.; Kalaichelvan, P.T. In vitro biological properties and characterization of nanosilver coated cotton fabrics—An application for antimicrobial textile finishing. *Int. Biodeterior. Biodegrad.* **2016**, *107*, 48–55. [CrossRef]
8. Botelho, C.M.; Fernandes, M.M.; Souza, J.M.; Dias, N.; Sousa, A.M.; Teixeira, J.A.; Fangueiro, R.; Zille, A. New textile for personal protective equipment—Plasma chitosan/silver nanoparticles nylon fabric. *Fibers* **2021**, *9*, 3. [CrossRef]
9. El-Rafie, M.; Ahmed, H.B.; Zahran, M. Characterization of nanosilver coated cotton fabrics and evaluation of its antibacterial efficacy. *Carbohydr. Polym.* **2014**, *107*, 174–181. [CrossRef]
10. Gadkari, R.R.; Ali, S.W.; Joshi, M.; Rajendran, S.; Das, A.; Alagirusamy, R. Leveraging antibacterial efficacy of silver loaded chitosan nanoparticles on layer-by-layer self-assembled coated cotton fabric. *Int. J. Biol. Macromol.* **2020**, *162*, 548–560. [CrossRef] [PubMed]
11. Petkova, P.; Francesko, A.; Perelshtein, I.; Gedanken, A.; Tzanov, T. Simultaneous sonochemical-enzymatic coating of medical textiles with antibacterial ZnO nanoparticles. *Ultrason. Sonochemistry* **2016**, *29*, 244–250. [CrossRef] [PubMed]
12. Zheng, Y.; Xiao, M.; Jiang, S.; Ding, F.; Wang, J. Coating fabrics with gold nanorods for colouring, UV-protection, and antibacterial functions. *Nanoscale* **2013**, *5*, 788–795. [CrossRef]
13. El-Naggar, M.E.; Shaarawy, S.; Hebeish, A. Bactericidal finishing of loomstate, scoured and bleached cotton fibres via sustainable in-situ synthesis of silver nanoparticles. *Int. J. Biol. Macromol.* **2018**, *106*, 1192–1202. [CrossRef]
14. Shcherbakov, A.B.; Reukov, V.V.; Yakimansky, A.V.; Krasnopeeva, E.L.; Ivanova, O.S.; Popov, A.L.; Ivanov, V.K. $CeO_2$ nanoparticle-containing polymers for biomedical applications: A review. *Polymers* **2021**, *13*, 924. [CrossRef]
15. Qian, J.; Dong, Q.; Chun, K.; Zhu, D.; Zhang, X.; Mao, Y.; Culver, J.N.; Tai, S.; German, J.R.; Dean, D.P.; et al. Highly stable, antiviral, antibacterial cotton textiles via molecular engineering. *Nat. Nanotechnol.* **2023**, *18*, 168–176. [CrossRef]
16. Gulati, R.; Sharma, S.; Sharma, R.K. Antimicrobial textile: Recent developments and functional perspective. *Polym. Bull.* **2022**, *79*, 5747–5771. [CrossRef]
17. Li, Y.; Miao, Q.; Wang, X. Antibacterial capability of air filter fiber materials treated with triclosan against indoor environmental microbes. *Atmosphere* **2022**, *13*, 1104. [CrossRef]
18. Shrestha, P.; Zhang, Y.; Chen, W.J.; Wong, T.Y. Triclosan: Antimicrobial mechanisms, antibiotics interactions, clinical applications, and human health. *J. Environ. Sci. Health Part C* **2020**, *38*, 245–268. [CrossRef]
19. Qian, L.; Sun, G. Durable and regenerable antimicrobial textiles: Improving efficacy and durability of biocidal functions. *J. Appl. Polym. Sci.* **2004**, *91*, 2588–2593. [CrossRef]
20. Ma, Y.; Aslam, M.Z.; Wu, M.; Nitin, N.; Sun, G. Strategies and perspectives of developing anti-biofilm materials for improved food safety. *Food Res. Int.* **2022**, *159*, 111543. [CrossRef] [PubMed]
21. Sun, X.; Zhang, L.; Cao, Z.; Deng, Y.; Liu, L.; Fong, H.; Sun, Y. Electrospun composite nanofiber fabrics containing uniformly dispersed antimicrobial agents as an innovative type of polymeric materials with superior antimicrobial efficacy. *ACS Appl. Mater. Interfaces* **2010**, *2*, 952–956. [CrossRef]
22. Saverina, E.A.; Frolov, N.A.; Kamanina, O.A.; Arlyapov, V.A.; Vereshchagin, A.N.; Ananikov, V.P. From Antibacterial to Antibiofilm Targeting: An Emerging Paradigm Shift in the Development of Quaternary Ammonium Compounds (QACs). *ACS Infect. Dis.* **2023**, *9*, 394–422. [CrossRef]
23. Wang, P.; Zhang, M.; Qu, J.; Wang, L.; Geng, J.; Fu, F.; Liu, X. Antibacterial cotton fabric prepared by a "grafting to" strategy using a QAC copolymer. *Cellulose* **2022**, *29*, 3569–3581. [CrossRef]
24. Yueh, M.-F.; Tukey, R.H. Triclosan: A widespread environmental toxicant with many biological effects. *Annu. Rev. Pharmacol. Toxicol.* **2016**, *56*, 251–272. [CrossRef]
25. Reshma, A.; Priyadarisini, V.; Amutha, K. Sustainable antimicrobial finishing of fabrics using natural bioactive agents—A review. *Int. J. Life Sci. Pharma Res.* **2018**, *8*, 10–20. [CrossRef]

26. Yıldırım, F.F.; Avinc, O.; Yavas, A.; Sevgisunar, G. Sustainable antifungal and antibacterial textiles using natural resources. In *Sustainability in the Textile and Apparel Industries: Sourcing Natural Raw Materials*; Springer: Cham, Switzerland, 2020; pp. 111–179.
27. Tanasa, F.; Teaca, C.-A.; Nechifor, M.; Ignat, M.; Duceac, I.A.; Ignat, L. Highly Specialized Textiles with Antimicrobial Functionality—Advances and Challenges. *Textiles* **2023**, *3*, 219–245. [CrossRef]
28. Vertuccio, L.; Guadagno, L.; D'angelo, A.; Viola, V.; Raimondo, M.; Catauro, M. Sol-gel synthesis of caffeic acid entrapped in silica/polyethylene glycol based organic-inorganic hybrids: Drug delivery and biological properties. *Appl. Sci.* **2023**, *13*, 2164. [CrossRef]
29. Catauro, M.; D'Angelo, A.; Fiorentino, M.; Pacifico, S.; Latini, A.; Brutti, S.; Ciprioti, S.V. Thermal, spectroscopic characterization and evaluation of antibacterial and cytotoxicity properties of quercetin-PEG-silica hybrid materials. *Ceram. Int.* **2023**, *49*, 14855–14863. [CrossRef]
30. Tan, L.; Sin, L.T.; Bee, S.; Ratnam, C.T.; Woo, K.; Tee, T.; Rahmat, A.R. A review of antimicrobial fabric containing nanostructures metal-based compound. *J. Vinyl Addit. Technol.* **2019**, *25*, E3–E27. [CrossRef]
31. Sfameni, S.; Hadhri, M.; Rando, G.; Drommi, D.; Rosace, G.; Trovato, V.; Plutino, M.R. Inorganic finishing for textile fabrics: Recent advances in wear-resistant, UV protection and antimicrobial treatments. *Inorganics* **2023**, *11*, 19. [CrossRef]
32. Feng, Q.L.; Wu, J.; Chen, G.Q.; Cui, F.Z.; Kim, T.N.; Kim, J.O. A mechanistic study of the antibacterial effect of silver ions on Escherichia aureus and Staphylococcus aureus. *J. Biomed. Mater. Res.* **2000**, *52*, 662–668. [CrossRef]
33. Yin, I.X.; Zhang, J.; Zhao, I.S.; Mei, M.L.; Li, Q.; Chu, C.H. The antibacterial mechanism of silver nanoparticles and its application in dentistry. *Int. J. Nanomed.* **2020**, *15*, 2555–2562. [CrossRef] [PubMed]
34. Bektas, N.; Şenel, B.; Yenilmez, E.; Özatik, O.; Arslan, R. Evaluation of wound healing effect of chitosan-based gel formulation containing vitexin. *Saudi Pharm. J.* **2020**, *28*, 87–94. [CrossRef]
35. Sánchez, R.; Alonso, G.; Valencia, C.; Franco, J. Rheological and TGA study of acylated chitosan gel-like dispersions in castor oil: Influence of acyl substituent and acylation protocol. *Chem. Eng. Res. Des.* **2015**, *100*, 170–178. [CrossRef]
36. Mohammadi, H.; Alihosseini, F.; Hosseini, S.A. Improving physical and biological properties of nylon monofilament as suture by Chitosan/Hyaluronic acid. *Int. J. Biol. Macromol.* **2020**, *164*, 3394–3402. [CrossRef]
37. Kumar-Krishnan, S.; Prokhorov, E.; Hernández-Iturriaga, M.; Mota-Morales, J.D.; Vázquez-Lepe, M.; Kovalenko, Y.; Sanchez, I.C.; Luna-Bárcenas, G. Chitosan/silver nanocomposites: Synergistic antibacterial action of silver nanoparticles and silver ions. *Eur. Polym. J.* **2015**, *67*, 242–251. [CrossRef]
38. Grgac, S.F.; Tarbuk, A.; Dekanić, T.; Sujka, W.; Draczyński, Z. The chitosan implementation into cotton and polyester/cotton blend fabrics. *Materials* **2020**, *13*, 1616. [CrossRef]
39. El-tahlawy, K.F.; El-Bendary, M.A.; Elhendawy, A.G.; Hudson, S.M. The antimicrobial activity of cotton fabrics treated with different crosslinking agents and chitosan. *Carbohydr. Polym.* **2005**, *60*, 421–430. [CrossRef]
40. Cao, X.L.; Cheng, C.; Ma, Y.L.; Zhao, C.S. Preparation of silver nanoparticles with antimicrobial activities and the researches of their biocompatibilities. *J. Mater. Sci. Mater. Med.* **2010**, *21*, 2861–2868. [CrossRef] [PubMed]
41. Wei, D.; Sun, W.; Qian, W.; Ye, Y.; Ma, X. The synthesis of chitosan-based silver nanoparticles and their antibacterial activity. *Carbohydr. Res.* **2009**, *344*, 2375–2382. [CrossRef]
42. Rezazadeh, N.H.; Buazar, F.; Matroodi, S. Synergistic effects of combinatorial chitosan and polyphenol biomolecules on enhanced antibacterial activity of biofunctionalized silver nanoparticles. *Sci. Rep.* **2020**, *10*, 19615. [CrossRef] [PubMed]
43. Nithya, A.; JeevaKumari, H.L.; Rokesh, K.; Ruckmani, K.; Jeganathan, K.; Jothivenkatachalam, K. A versatile effect of chitosan-silver nanocomposite for surface plasmonic photocatalytic and antibacterial activity. *J. Photochem. Photobiol. B Biol.* **2015**, *153*, 412–422. [CrossRef] [PubMed]
44. Mollick, S.; Islam, T.; Miah, R.; Riad, M.H.; Chakrabarty, T.; Hossen, T.; Islam, T. Development of Antibacterial Cotton Fabric Utilizing Microencapsulation Technique from Dragon Fruit Peel Extract. *Fibers Polym.* **2023**, *24*, 3937–3945. [CrossRef]
45. Safi, M.; Gheisar, F.; Najafi, F.; Hadavand, B.S. Investigation of Shade Darkening Property of Polydimethylsiloxane and Poly (Dimethyl/Diphenyl Siloxane) Modified with Amino Groups on Dyed Textiles. *Fibers Polym.* **2023**, *24*, 4213–4227. [CrossRef]
46. Emam, H.E.; Rehan, M.; Mashaly, H.M.; Ahmed, H.B. Large scaled strategy for natural/synthetic fabrics functionalization via immediate assembly of AgNPs. *Dye. Pigment.* **2016**, *133*, 173–183. [CrossRef]
47. Rai, S.; Saremi, R.; Sharma, S.; Minko, S. Environment-friendly nanocellulose-indigo dyeing of textiles. *Green Chem.* **2021**, *23*, 7937–7944. [CrossRef]