# Ultrasound Radiation as a "Throwing Stones" Technique for the Production of Antibacterial Nanocomposite Textiles

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**ABSTRACT** Ultrasound irradiation was applied as a "throwing stones" technique for coating cotton bandages with MgO and  $Al_2O_5$  commercially obtained nanoparticles. The homogeneous distribution of the nanoparticles without any aggregation on the fabrics was demonstrated. The antibacterial activities of the MgO/Al<sub>2</sub>O<sub>3</sub>—fabric nanocomposite were tested against *Escherichia coli* (Gram negative) and *Staphylococcus aureus* (Gram positive) cultures. A significant bactericidal effect, even in a concentration <1 % (by weight), was detected.

**KEYWORDS:** MgO • Al<sub>2</sub>O<sub>3</sub> • nanoparticles • ultrasound irradiation • antimicrobial activity • coating

# 1. INTRODUCTION

The science and technology of nanoobjects with dimensions <100 nm present both basic challenges and enormous potential for applications (1). Research in the area of nanometer-scaled metal oxides has shown encouraging discoveries that have demonstrated a clear size dependence of their electromagnetic, optical, and catalytic properties (2, 3). One striking characteristic of nanometerscaled metal oxides is their broad range of probable reactions with the biological cells in general and with bacterial cells in particular (4, 5).

Since the introduction of penicillin in the 1940s, antibiotics had become the standard treatment for bacterial infections. However, during the past decades, bacteria have possessed one of the most worrisome characteristics, i.e., evolving the ability to withstand the effects of antibiotics (6). Such a trait, termed "antibiotic resistance", encompasses also organisms other than bacteria, e.g., fungi (7). Therefore, metal oxide nanoparticles (NPs), as antimicrobial agents, can fulfill the need of a new class of biocidal formulations. Moreover, metal oxide NPs have the key advantage that they are inorganic materials and, therefore, hold the ability to withstand adverse processing conditions (8). Some metal nanooxides, such as aluminum oxide (Al<sub>2</sub>O<sub>3</sub>), titanium oxide (TiO<sub>2</sub>), and magnesium oxide (MgO), are nontoxic for the human body, and during the past decade, intensive research involving metal oxide NPs was intensified, focusing on the production of textiles with antibacterial functions (9).

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 $Al_2O_3$  NPs have important applications in the ceramics industry (10) and can be used as an an abrasive material, as anabsorbent, as a biomaterial, as reinforcements of metal-matrix composites, and in heterogeneous catalysis (11, 12).

There have been very few studies reported in the literature on the interaction of alumina NPs with microbes. The studies of the antibacterial properties of alumina showed no detrimental effect of an alumina slurry within the 62.5–250 mg/L concentration range on *Escherichia coli* (13). One of the objectives of the present work was to investigate the possible growth-inhibitory effect of alumina-coated fabrics on environmentally relevant Gram-negative and Gram-positive model microorganisms.

MgO is well-known to have a strong antibacterial activity (14-17). Different methods were reported on the synthesis of MgO NPs, such as the controlled speed of formation following the heating procedure (14), microwave-assisted synthesis (16), formation from aqueous droplets in a flame spray pyrolysis reactor (18), and sonochemically enhanced hydrolysis followed by supercritical drying (19).

To date, there is still a lack of definite knowledge regarding the interaction of metal oxide NPs with the bacterial cell. For MgO, as well as for ZnO, some researchers have manifested the role of reactive oxygen species (ROS) generated on the wet oxide surfaces due to the reactive nature of the surface defect sites (16, 20). In this sense, the high surfaceto-volume ratio of the nanometer scale of the oxides results in the formation of more ROS per unit weight. Thus, the bacterial cell architecture is impaired by the detrimental effect of the increased amount of ROS (20-22). In regards to the nanometer scale of Al<sub>2</sub>O<sub>3</sub>, there is a paucity of prior literature reports on its effect on bacterial species; for example, Sadiq et al. suggested that a mechanism of strong

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adhesion of the alumina NPs to the *E. coli* bacteria cells, derived from electrostatic forces, is a critical factor underlying the alumina NPs antibacterial activity toward *E. coli* (23). However, the exact toxicity mechanism is yet to be discerned.

Research involving nanotechnology for improving the performance or creating unprecedented functions of textile materials is flourishing. The impact of nanotechnology in the textile industry has made it possible to produce a new generation of antimicrobial textiles by innovative finishes of the fabric's surface. Gabbay and co-workers have demonstrated the finishing of cotton and polyester fibers with 10 wt % CuO using a multiphase soaking procedure (24). There are only a few methods in the literature that describe the coating of fabrics with ZnO NPs, e.g., the "pad-dry-cure" method (25), radiation, and thermal treatments (26). These techniques have some disadvantages: first, they are rather complicated and involve several stages, and second, a stabilizing agent is used in order to get smaller ZnO NPs, resulting in the presence of impurities in the final products. As far as we know, there are no publications on the production and application of the MgO and Al<sub>2</sub>O<sub>3</sub> textile composite.

Sonochemical irradiation has been proven as an effective technique for the synthesis of nanophased materials (27), as well as for the deposition and insertion of NPs on/into mesoporous ceramic and polymer supports, fabrics, and glass (28-30). The mechanism of deposition involves microjets formed after the collapse of the acoustic bubble. These microjets push the newly formed NPs onto the substrate surface at such a high speed that it causes their embedment in the substrate. In our previous investigation (31), we demonstrated the excellent antibacterial activity of ZnO NPs uniformly deposited onto the surface of a cotton bandage by the sonochemical method. The deposition of ZnO on cotton bandages was performed in an in situ reaction; namely, the NPs were formed and simultaneously deposited on the fabric's surface. The deposition of MgO and Al<sub>2</sub>O<sub>3</sub> NPs on fabrics in a one-step sonochemical reaction is impossible. That is because the sonication of metal (M) acetate (where M = Mg, Al) leads to an amorphous phase of the corresponding M hydroxide and not the corresponding metal oxide. Antibacterial tests toward Staphylococcus aureus and *E. coli* show that these M hydroxides are not active as bactericides. Only the heating of the amorphous product up to several hundred degrees allows the formation of crystalline nanooxides. This action is not possible when the NPs are deposited on textiles because at such temperatures the textiles will be destroyed.

In the current study, ultrasound irradiation was applied as a "throwing stones" technique for deposition of MgO and  $Al_2O_3$  on the textile; namely, we used commercial nanopowders of MgO (<50 nm) and  $Al_2O_3$  (<50 nm) and sonicated them in the presence of the cotton fabric. The morphology of the commercial NPs of MgO and  $Al_2O_3$  was studied by high-resolution transmission electron microscopy (HRTEM) and reveals particles of sizes ranging from 20 to 45 nm.

The antibacterial properties of the MgO- and  $Al_2O_3$ -coated cottons against the Gram-negative *E. coli* and the Gram-

positive *S. aureus* bacteria strains were demonstrated. In addition, the current research shows that, even with the low loading of metal oxide (<1 wt %) on the cotton, an effective antibacterial activity was detected.

# 2. EXPERIMENTAL SECTION

**2.1.** Deposition of MgO/Al<sub>2</sub>O<sub>3</sub> on the Fabric. All of the chemical reagents, of chemical grade, were purchased from Aldrich and used without further purification. The sonication was conducted in the presence of a cotton bandage, with a 13 treads/cm<sup>2</sup> density. The reaction parameters were varied to obtain the best conditions for coating of the NPs on the bandages. The optimal results representing a typical experiment were as follows: 1 piece of  $10 \times 10$  cm bandage (0.7 g) and 0.8 g of MgO/Al<sub>2</sub>O<sub>3</sub> NPs were added to a solution of 4:1 ethanol/ ethylene glycol (EG) in a 100 mL sonication flask. The reaction mixture was irradiated for 2 h with a high-intensity ultrasonic horn (Ti-horn, 20 kHz, 750 W at 70% efficiency). At the end of the procedure, the color of the fabric had not changed. The product was washed thoroughly with ethanol and dried under vacuum.

2.2. Antimicrobial Test. The antimicrobial activity of the MgO- or Al<sub>2</sub>O<sub>3</sub>-coated bandages was tested against the Gramnegative bacterium E. coli and the Gram-positive bacterium S. aureus following a AATCC standard procedure (test method 100). The coated bandages were sterilized before checking the antibacterial activity. Cultures of the bacteria were grown overnight in an NP medium at 37 °C with aeration and transferred the next morning into a fresh medium at an initial optical density (OD) of 0.1 at 600 nm. When the culture reached an OD of 0.3 (600 nm) in the log phase, the cells were harvested by centrifugation and washed twice with a 0.9% solution of NaCl at pH 6.5. The cells were then diluted with a 0.9% solution of NaCl. A total of 1 cm<sup>2</sup> each of the coated and uncoated fabrics, which served as references was placed in a vial containing 4.5 mL of a 0.9% NaCl solution. A total of 500  $\mu$ L of the washed and diluted cells was transferred into the vial. The initial cell number in the vial was approximately 10<sup>7</sup> CFU/mL. To ensure that any decrease in the bacterial number was likely to be due to exposure to a coated bandage treatment, a 0.9% solution of NaCl without any fabric was included in the experiment as an additional control. The bacterial suspensions were incubated for up to 2 h at 37 °C and 170 rpm. Samples of 100  $\mu$ L each were taken at the beginning, after 60 min, and after 2 and 3 h. Different dilutions were transferred onto nutrient agar plates. The plates were allowed to grow overnight at 37 °C and then counted for viable bacteria. The viable bacteria were monitored by counting the number of colony-forming units (CFUs) from the appropriate dilution on nutrient agar plates. The experiments were repeated at least three times for each bacterium. All experiments were recorded on an Eppendorf Biophotometer.

2.3. Optimization of the Reaction Conditions. One of the aims of this research was to reach a minimal effective concentration of the deposited metal oxide NPs on the fabrics, which would still demonstrate antibacterial activity. The current research probes the hypothesis that ultrasound can also be used for a two-stage coating using commercial NPs for deposition. We hypothesize that the "throwing stones" technique will succeed because microjets will push the commercial NP in the same way that it throws the sonochemically formed NP onto the fabric. Our previous studies indicated that the yield of the product and the particles' sizes are strongly dependent on the rate of particle collision, on the concentration of the reagents, and on the reaction medium during sonochemical synthesis (31). That is why experimental parameters such as time and the amounts of the precursor and solvent were selected as important factors for optimization of the sonochemical reaction.

# Table 1. Optimization of the MgO Coating Process

expt	solvent/weight of MgO (g)	reaction time (h)	content of MgO (wt %)	antibacterial activity (% reduction)
1	water 0.4	2		
2	water/ethanol <sup>a</sup> 0.4	1	0.23	
3	water/ethanol <sup>a</sup> 0.4	2	0.27	
4	water/EG <sup>b</sup> 0.4	2	0.32	
5	water/EG <sup>b</sup> 0.8	2	0.36	
6	ethanol/EG <sup>c</sup> 0.8	1	0.52	55
7	ethanol/EG <sup>c</sup> 0.8	2	0.8	99.9

<sup>*a*</sup> 70 mL of water + 30 mL of ethanol. <sup>*b*</sup> 70 mL of water + 30 mL of EG. <sup>*c*</sup> 80 mL of ethanol + 20 mL of EG.

The different compositions used in the reaction are presented in Table 1. The initial experiments on the sonochemical deposition of MgO nanopowder on the fabric were carried out in the most environmentally friendly solvent: water (expt 1). The titration analysis of the fabric after reaction does not reveal any presence of MgO. Changing the amount of the precursor and/ or the reaction time does not result in an increase of the amount of the deposited metal oxide. In the next experiment, the deposition was performed in a water/ethanol solution. The commercial nanopowder of MgO was sonicated in the presence of a fabric in a water/ethanol (70:30) solution for 1 h (expt 2). The quantitative analysis indicated that the amount of deposited material was very low, and no antibacterial effect was detected. Even after an increase in the reaction time to 2 h (expt 3), the amount of deposited material was poor. In expt 4, EG was added instead of ethanol, but the concentration of deposited MgO did not change significantly. Increasing the initial amount of MgO (expt 5) in the same reaction medium as that in expt 4 did not influence the quantity of the deposited material. The reason for the poor coating in water solutions is the partial hydrolysis of MgO. Using pure ethanol or EG in sonochemical reactions is not effective because of the fact that the ethanol has a high vapor pressure, which prevents the creation of acoustic bubbles, and that EG is too viscous for ultrasound irradiation. The next experiments were conducted in an ethanol/EG (4:1) solution for 1 h (expt 6). The amount of deposited MgO increased, but a very poor antibacterial effect was detected. Increasing the reaction time to 2 h (expt 7) led to a larger amount of deposited MgO, and an excellent antibacterial effect was obtained. The optimal reaction conditions were found in expt 7. Thus, this sample was chosen for further investigations. These conditions were also used for deposition of  $Al_2O_3$ , and the amount of coating was found to be 1 wt %.

We have also tried to deposit the MgO and  $Al_2O_3$  NPs on the fabric without using ultrasonic waves and failed to obtain any coating.

#### **3. CHARACTERIZATION**

The MgO content on the fabrics was determined by volumetric titration with ethylenediaminetetraacetic acid after dissolution of the sonicated product in  $0.5 \text{ N HNO}_3$  and controlled by inductively coupled plasma (ICP) analysis on the device ULTIMA JY2501. The Mg concentration was found to be 0.8 wt % MgO.

The  $Al_2O_3$  content on the bandage was determined by elemental analysis of the pristine and coated fabric with elemental analyzers (Flash EA 1112). The cotton fabric comprises carbon, hydrogen, and oxygen, and its thermal decomposition provides information about the content of each element when the total is 100%. The measured element percentages of carbon, hydrogen, and oxygen in the uncoated fabric were 43.42, 6.54, and 49.99 wt %, respectively, (total: 99.95 wt %). The percentage of the elements in the coated fabric was measured as 49.19 wt % carbon, 6.79 wt % hydrogen, and 42.96 wt % oxygen (total: 98.94 wt %). The remainder,  $\sim$ 1 wt %, is related to the content of Al<sub>2</sub>O<sub>3</sub> in the composite.

The particles' morphology and size were studied with a high-resolution scanning electron microscope JEOL-JSN 7000F and with a high-resolution transmission electron microscope JEOL-2100, working at 200 kV accelerating voltage. The coated textile sample was prepared for electron microscopy by the epoxy embedding technique following ultramicrotome cutting. The X-ray diffraction (XRD) pattern of the MgO nanopowder was measured with a Bruker D8 diffractometer (Karlsruhe, Germany) with Cu K $\alpha$  radiation. The presence of NPs in the washing solution was tested by HRTEM and by dynamic light scattering (DLS) measurements, employing a Coulter particle analyzer instrument (N4 plus). The tests were conducted in a washing solution after soaking the bandage in 0.9% NaCl for 96 h at 37 °C. The mechanical tests were performed on a mechanical testing machine, Zwick 1445 (Zwick Gmbh & Co., Ulm-Einsingen, Germany).

### 4. RESULTS AND DISCUSSION

4.1. Structure and Morphology of the MgO/ $Al_2O_3$ -Fabric Nanocomposite. In order to study the morphology of the  $Al_2O_3$ -coated fabric and the influence of the ultrasound on the structure of the NPs, a high-resolution transmission electron microscope was used. The cotton fibers were embedded in the copper grid, using the epoxy-embedding technique and ultramicrotome cutting.

Before the sonochemical coating reaction, the commercial Al<sub>2</sub>O<sub>3</sub> nanopowder was tested by XRD and a single crystalline phase of orthorhombic  $\delta$ -Al<sub>2</sub>O<sub>3</sub> was detected (PDF No. 046-1215). After the reaction, HRTEM of the Al<sub>2</sub>O<sub>3</sub>coated fabric is depicted in Figure 2a and a uniform distribution of the particles along the fiber is observed. The redmarked particle in Figure 1 a was taken under high magnification (Figure 1b), which provides further verification for identification of the coating as  $Al_2O_3$ . The measured distances between (131), (220), and (311) lattice planes are 0.25, 0.28, and 0.25 nm, respectively, which match very well the distances reported in the literature for the orthorhombic lattice of  $\delta$ -Al<sub>2</sub>O<sub>3</sub> (PDF No. 046-1215). These results indicate that ultrasound irradiation can be used as an effective method for coating textiles without causing any damage to the crystalline structure of the commercial NPs.

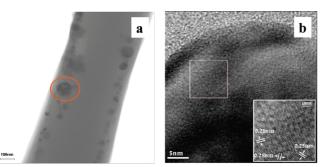


FIGURE 1. (a) HRTEM of a  $Al_2O_3\text{-}coated$  bandage. (b) High magnification ( $\times1\text{M})$  of the red-marked particle.

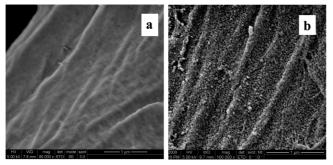


FIGURE 2. SEM images of (a) a pristine cotton bandage and (b) a MgO-coated bandage.

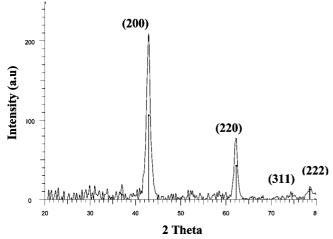


FIGURE 3. XRD pattern of MgO NPs after sonication.

The morphology of the fiber surface before and after the deposition of MgO NPs studied by high-resolution scanning electron microscopy (HRSEM) is presented in Figure 2. The image in Figure 2a demonstrates the smooth structure of the cotton fabric before coating with MgO NPs. After sonication, the homogeneous deposition of NPs on the cotton yarn is observed (Figure 2b). The particle size is in the low nanometric range ( $\sim$ 20–30 nm). The NPs are well dispersed along the fibers; however, some aggregation can be found.

In order to confirm that the structures of the MgO NPs are not damaged during sonication, a control experiment was carried out; namely, the MgO nanopowder was sonicated under the same reaction conditions as those of experiment 7, and at the end of the reaction; the powder was centrifuged and dried. The XRD patterns (Figure 3) of sonochemically treated MgO NPs demonstrate that MgO is crystalline in nature, and the diffraction peaks match a cubic

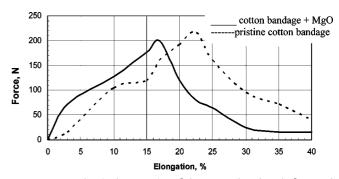


FIGURE 4. Mechanical properties of the cotton bandage before and after the deposition of MgO NPs.

phase of MgO (PDF No. 004-0829). The peaks at  $2\theta = 42.9$ , 62.3, 74.67, and 78.61° are assigned to the (200), (220), (311), and (222) reflection lines of cubic MgO particles, respectively. No peaks characteristic of any impurities were detected.

**4.2.** Mechanical Properties. The tensile mechanical properties of a common impregnated fabric were studied on a universal testing machine, Zwick 1445. Four-folded fabric samples with gauge lengths of 60 mm and widths of 40 mm were placed in special grips. The MgO-coated samples showed a rather more brittle behavior compared to the pristine bandage. Ultimately, the tensile strength for the coated sample was ~10% less than that of the nonimpregnated piece (Figure 4). The same behavior was obtained for a bandage coated with  $Al_2O_3$  NPs. The observed change in the mechanical behavior of the yarn is in a range that is acceptable for standard cotton fabrics. According to this result, we conclude that the sonochemical treatment of the fabric did not cause any significant damage to the structure of the yarn.

4.3. "Throwing Stones" Mechanism of Sonochemical Coating. In previous experiments (31), we have deposited NPs that were obtained in the sonochemical reaction on fabrics. In the current case, the NPs were not synthesized in the reaction but purchased from commercial sources. The following is a proposed mechanism for the strong adherence of the NPs to the fabric. When cavitation occurs in a liquid near a solid surface, the dynamics of the cavity collapse change dramatically. In pure liquids, the cavity remains spherical during collapse because its surroundings are uniform. However, close to a solid boundary, cavity collapse is very asymmetric and generates high-speed jets of the liquid. The potential energy of the expanded bubble is converted into the kinetic energy of a liquid jet that moves through the bubble's interior and penetrates the opposite bubble wall. These jets hit the surface with a tremendous force. When the suspension of the preliminary synthesized NPs and the fabric is irradiated, the microjets formed after the collapse of the bubble throw the NPs as "stones" at high speed to the cotton varns. It is possible, as suggested before (31), that a local melting of the fabric occurs when the particles impinge on the fabric. The proposed "throwing stones" mechanism for the sonochemical deposition of NPs is schematically presented in Figure 5. Such a use of ultrasound irradiation for coating processes

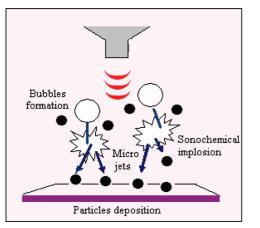


FIGURE 5. Scheme of the sonochemical deposition of NPs on the solid substrate.

differs from our previous reports where the NPs of CuO and ZnO were formed from the precursor solution under the ultrasound irradiation and, subsequently, thrown to the solid surface by the microjets. The main distinction between the one and two stages is the measured amount of the deposited material. The anchored amount for the commercial NP was found to be smaller by a factor of 2-3, as compared to the one-stage deposition, because of the fact that not all of the particles are pushed by the microjets to the fabric's surface. Nevertheless, using ultrasound as a "throwing stones" technique is an efficient process, especially for NPs that cannot be synthesized sonochemically.

**4.4. Bactericidal Tests.** The antibacterial activity of cotton fabrics coated with 0.8% MgO was determined using the Gram-positive bacterium *S. aureus* and the Gram-negative bacterium *E. coli*. As shown in Table 2, treatment for 1 h with the MgO-coated cotton leads to a complete growth inhibition of *E. coli*. Regarding *S. aureus*, a 100% reduction in viability was reached after 3 h, while after 1 h of treatment, a reduction of 90% could be seen. As for Al<sub>2</sub>O<sub>3</sub>, it is evident from this study that alumina NPs possess milder antimicrobial properties. Treatment for 1 h with the Al<sub>2</sub>O<sub>3</sub>-coated cotton leads to about a 23% growth inhibition of *E.* 

*coli* and an 84% growth after 3 h. Regarding *S. aureus*, only an 11% growth inhibition was observed after 1 h of treatment and about 75% after 3 h. Experiments of MgO- and Al<sub>2</sub>O<sub>3</sub>-coated bandages toward *E. coli* and *S. aureus* were repeated five times. Statistical interpretation for MgO experiment-killing results (in percentage). The mean values for *E. coli* after treatment for 60 min, 99.80  $\pm$  0.2, and 180 min, 99.96  $\pm$  0.058. The results for *E. coli* are reproducible; the same is true for *S. aureus*. The mean values for *S. aureus* elimination: 60 min, 89.244  $\pm$  0.8566; 180 min, 99.99512  $\pm$  0.0068. If we compare the results for the two strains, the difference between the two strains after 60 min is highly significant,  $P \leq$  0.005. After 180 min, the difference is for sure not significant.

Statistical interpretation for  $Al_2O_3$  experiments. The mean values for *E. coli*: 60 min, 23.28 ± 0.1568; 180 min, 83.85 ± 0.823. The mean values for *S. aureus*: 60 min, 11.05 ± 0.068; 180 min, 74.9477 ± 0.0023.

We suggest that the cytototoxic effect of  $Al_2O_3$  differs from MgO in its mechanism; i.e., electrostatic positive charges of the alumina are responsible for bacterial membrane disruption rather than ROS production, as assumed in the case of MgO (23).

# 5. LEACHING OF THE NPS

One of the factors influencing the commercial exploitation of the antibacterial fabrics is the release of the NPs into the surrounding environment.

The main hazard arises from the release of the MgO/Al<sub>2</sub>O<sub>3</sub> NPs into water upon laundering. To examine the leaching of the NPs, we have conducted some control experiments. Namely, we treated the coated bandages with an aqueous solution of sodium chloride. We placed a piece (0.08 g) of the coated bandage into 50 mL of a 0.9% NaCl solution at 37 °C for 96 h. The methods we used for revealing the presence of NPs were DLS and TEM. The results indicate that there is no presence of NPs in the washing solution after 96 h. That means that the sonochemically deposited NPs are strongly anchored to the textile substrate. The full explana-

Table 2.	Antibacterial	Activity	Tests	Using E.	coli and S.	aureus <sup>a</sup>

	duration of treatment							
	1 h			3 h				
treatment	$CFU, mL^{-1}$	$N/N_0$	% reduction in viability	$CFU, mL^{-1}$	$N/N_0$	% reduction in viability		
			E. coli					
clean fabric	$6.2 \times 10^{6}$	0.95	4.6	$6.39 \times 10^{6}$	0.98	1.7		
no fabric	$5.8 \times 10^{6}$	1.05	-5.5	$5.09 \times 10^{6}$	0.925	7.5		
MgO	$5.5 \times 10^{3}$	$8.1 \times 10^{-4}$	99.9	260	$\sim 3.8 \times 10^{-5}$	100.00		
$Al_2O_3$	$5.0 \times 10^{6}$	0.77	23.43	$1.0 \times 10^{5}$	0.16	84.10		
			S. aureus					
clean fabric	$6.5 \times 10^{6}$	0.87	13.33	$5.7 \times 10^{6}$	0.67	24		
no fabric	$6.7 \times 10^{6}$	0.94	5.63	$6.5 \times 10^{6}$	0.87	13.33		
MgO	$6.3 \times 10^{5}$	$9.5 \times 10^{-2}$	90.45	$2.58 \times 10^{3}$	$3.9 \times 10^{-4}$	99.96		
Al <sub>2</sub> O <sub>3</sub>	$6.3 \times 10^{6}$	0.89	11.12	$1.76 \times 10^{6}$	0.25	74.95		

<sup>*a*</sup> The viable bacteria were monitored by counting the number of colony-forming units (CFU);  $N/N_0$  = survival fraction.

tion for this phenomenon is described in detail in section 4.3. The microjets formed after the collapse of the bubble throw the NPs as "stones" at high speed to the yarns.

While no presence of NPs was found in the washing solution, leaching of the  $Mg^{2+}$  or  $Al^{3+}$  ions can occur during exposure of the coated fabrics to a wet environment. The content of Mg<sup>+2</sup> in the solution was determined by ICP and estimated to be 1.2 ppm. In order to examine the influence of Mg ions on the antibacterial effect, we performed an antibacterial test using a supernatant solution instead of the coated bandage. After incubation for 24 h at 37 °C, no reduction in E. coli after 24 h was observed. This result indicates that the  $Mg^{2+}$  ions have no influence on the antibacterial activity. The explanation for the presence of  $Mg^{2+}$  ions is the fact that the leaching of ions is governed by the  $K_{sp}$  value of each material. The solubility of MgO is 0.086 g/L, and this means that a very small amount of Mg<sup>2+</sup> ions will be released from the coated fabrics if equilibrium is established and the concentration will be smaller if no equilibrium is achieved. The Mg <sup>2+</sup> ions are considered to be safe for the environment and humans. The presence of Al<sup>3+</sup> ions cannot be revealed by ICP because this material has a much smaller  $K_{sp}$  than that of MgO. Therefore, no presence of  $Al^{3+}$  ions is expected to be found in the washing solution.

# 6. CONCLUSION

MgO and Al<sub>2</sub>O<sub>3</sub> NPs were deposited on the surface of cotton fabrics by ultrasound irradiation. This research demonstrates the successful use of ultrasound as a "throwing stones" technique for coating textiles with antimicrobial NPs. This approach is simple and includes an environmentally friendly process without involving any toxic chemical reagents. The physical and chemical analysis has shown that nanocrystalline MgO and Al<sub>2</sub>O<sub>3</sub> is finely dispersed on the fabric's surface without any damage to its structure and no significant aggregation was observed. The performance of coated fabrics as antibacterial agents was investigated, and the killing effect of bacteria was demonstrated. A strong bactericidal effect was observed for the MgO-coated fabric. The antibacterial effect of the Al<sub>2</sub>O<sub>3</sub>-coated fabric was found to be less significant than that of MgO. This difference in the antibacterial mode of action of the two nanooxides is hypothesized to be derived from a difference in their antibacterial mechanisms. The coated fabrics can have potential applications in wound dressing and bed linen and as bandages. They can also be recommended for the purification of medical and food equipment, domestic cleaning, etc. We have already developed a small pilot line that can take 50 m of fabric with a 10 cm width and coat it roll-to-roll with NPs (32). The sonochemical process can be upscaled for larger dimensions, and such a machine can be built as a part of textile production.

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