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Topical Review

Application of nanotechnology in antimicrobial finishing of biomedical textiles

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Abstract

In recent years, the antimicrobial nanofinishing of biomedical textiles has become a very active, high-growth research field, assuming great importance among all available material surface modifications in the textile industry. This review offers the opportunity to update and critically discuss the latest advances and applications in this field. The survey suggests an emerging new paradigm in the production and distribution of nanoparticles for biomedical textile applications based on non-toxic renewable biopolymers such as chitosan, alginate and starch. Moreover, a relationship among metal and metal oxide nanoparticle (NP) size, its concentration on the fabric, and the antimicrobial activity exists, allowing the optimization of antimicrobial functionality.

Keywords: nanoparticles, antimicrobial, textile, zinc, silver, titanium, chitosan

Introduction

In the last 20 years, pathogenic bacteria have developed resistance to almost all the commercially available antibiotics, and the number of new antibiotics expected to enter the market is limited [1]. Thus, searching for new antibacterial agents is a priority for pharmaceutical companies and researchers. Recently, novel antimicrobial agents have been developed using nanoscale materials. Compared to classic antibiotics, these materials have a lower propensity to induce high-level, single-step resistance mutation due to their multi-targeted mechanism of action, high surface area to volume ratio, and unique chemical and physical

properties [2–4]. Numerous types of nanomaterials with antimicrobial properties such as copper [5, 6], zinc [7], titanium [8, 9], magnesium [10], gold [11], chitosan [12] and alginate [13] have been developed in recent years. However, among all, silver nanoparticles (AgNPs) have proved to be the most effective against bacteria, viruses and eukaryotic microorganisms and are being exploited in medicine for burn treatment, dental materials, metal coating, textile fabrics, water treatment and sunscreen lotions [1]. Moreover, silver has proved to have low toxicity to human cells, high thermal stability and low volatility [14]. It is known that the size, shape and crystalline structure of AgNPs affect their toxicological impact on microorganisms. However, the mechanism of bactericidal actions of AgNPs is still not fully elucidated, particularly because most of the available toxicity data are obtained in water or cell culture media, which do not reflect the complex environment inside living organisms [15]. Nowadays, the prevailing paradigm suggests a combination of various factors: (i) Nanoscaled direct interactions between NPs and cell membranes affect their permeability and are followed by a cascade of intracellular reactions, including DNA condensation; (ii) Silver ions reacting with thiol groups of cellular proteins interferes with the bacterial respiratory chain; (iii) Extracellular and intracellular generations of reactive oxygen species have resulted in membrane lipid and DNA damage [14]. Antimicrobial finishing of textiles for biomedical purposes has become an important area of research and one of the fastest growing sectors of the textile market. The global Antimicrobial Coatings Market's worth in 2012 is $1.5 billion and is estimated to reach $2.9 billion by 2018, growing at a compound annual growth rate of 11.8% from 2013 to 2018 under normal conditions [16, 17]. In general, the activity of antimicrobial finishes in textiles can be classified as biocidal or biostatic [18]. While the biocides include agents that kill microorganisms, the biostatics inhibit the microorganisms' growth. Antimicrobial textiles commonly use biocides, such as metal nanoparticles (or their salts), quaternary ammonium compounds, poly (hexamethylene biguanide), triclosan and chitosan, as active agents. These agents are either incorporated into the fibers during extrusion or attached to their fiber surface during finishing [19]. However, the definitions of 'bacteriostatic' and 'bactericidal' do not enclose *two* pure categories of antimicrobial agents that exclusively kill bacteria or that only inhibit growth. Within 18–24 h after the test, bactericidal agents usually fail to kill every organism, especially if the inoculum is large, and bacteriostatic agents kill some bacteria [20]. Moreover, the *in vitro* microbiological determination of an antibacterial agent in textiles is also influenced by growth conditions, bacterial density, test duration, extent of reduction in bacterial numbers, fabric shape, morphology and type of material. The most effective methods for testing the efficacy of fabrics that contain antimicrobials are the AATCC 100 and the AATCC 147 from the American Association of Textile Chemists and Colorists. The first test determines both the bacteriostatic activity as well as the bactericidal activity; the second test detects bacteriostatic activity of diffusible antimicrobial agents on treated textile materials by determining a zone of inhibition. However, during the preparation of the manuscript and the compilation of table 1, it became evident that there was a large variety of different tests that could be utilized to determine the fabrics' antimicrobial activity as well as a lack of information about the methodologies utilized to determine whether an antibacterial agent is bactericidal or bacteriostatic. Most antibacterial textiles are better described as potentially being both bactericidal and bacteriostatic, but in this review, they have been considered 'bactericidal' textiles only when able to kill more than 99.9% of the inoculum.

Currently, nanotechnology is considered the most promising technology for novel textile commercial applications since it allows the permanent and effective functionalization of

Table 1. Antimicrobial nanomaterials applied on textiles.

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
Acrylic					
[Ag]	1–7	Photocured Carboxymethyl Starch NPs	20* [*S. aureus*] 15* [*E. coli*]	1* [*S. aureus*] (15) 0.5* [*E. coli*] (15)	[127]
Bamboo					
[Ag]	<100	NPs grafted with acrylic acid	98.7 [*S. aureus*] 100 [*E. coli*]	84.9 [*S. aureus*] (50) 96.2 [*E. coli*] (50)	[128]
[CuO]	<100 [1 wt.%]	NPs grafted with acrylamide	100 [*S. aureus*] 99 [*E. coli*]	75 [*S. aureus*] (50) 75 [*E. coli*] (50)	[94]
[ZnO]	10 [14 μg g^{-1}]	NPs grafted with HSDA	99.1 [*S. aureus*] 99.9 [*E. coli*]	99 [*S. aureus*] (20) 98.9 [*E. coli*] (20)	[87]
Cellulose acetate					
[Ag]	3–16 [0.05 wt.%]	Electrospinned nanofibers containing NPs	99.9 [*S. aureus*] 99.9 [*E. coli*] 99.9 [*K. pneumonia*] 99.9 [*P. aeruginosa*]	n.a.	[104]
	21 [0.5 wt.%]	Electrospinned nanofibers containing NPs	99.9 [*S. aureus*] 99.9 [*E. coli*] 99.9 [*K. pneumonia*] 99.9 [*P. aeruginosa*]	n.a.	[129]
Cotton					
[Alginate/TSA]	99 [70 μg g^{-1}]	Colloid NPs impregnated on fabric	99.9 [*E. coli*] 99.9 [*S. aureus*]	99 [*E. coli*] (30)	[130]
[Ag]	0.65 [7 wt.%]	Colloid NPs padded on fabric	1.21* [*S. aureus*]	0 [*S. aureus*] (5)	[23]
	1–2 [0.8 wt.%]	Silica–silver core–shell particle deposited by Pad-dry-cure method	<1* [*E. coli*]	n.a.	[40]
	1.6 [10 μg g^{-1}]	Biosynthesized NPs impregnated on cotton	99 [*S. aureus*]	n.a.	[131]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
	2–5 [3.17 wt.%]	Colloid NPs synthetized and adsorbed on fabric	99.9 [*S. typhimurium*] 97 [*S. aureus*]	96 [*S. typhimurium*] (30) 93 [*S. aureus*] (30)	[98]
	2–12 [1215 $\mu g\ g^{-1}$]	Colloid NPs adsorbed by exhaustion method	6–7* [*E. coli*]	n.a.	[39]
	2–12 [385 $\mu g\ g^{-1}$]	Colloid NPs adsorbed by exhaustion method	4–5* [*E. coli*]	n.a.	[39]
	2–5 [30 $\mu g\ g^{-1}$]	Colloid NPs padded on fabric	99.9 [*S. aureus*] 99.9 [*K. pneumoniae*]	94.5 [*K. pneumoniae*] (20) 97.2 [*S. aureus*] (20)	[132]
	2–6	NPs–poly(acrylate) clusters impregnated on fabric	>1* [*S. aureus*] >1* [*S. epidermidis*] >1* [*P. aeruginosa*] >1* [*C. albicans*]	n.a.	[44]
	2–8 [0.7 wt.%]	Cellulose–Gum polymer–Ag nanocomposite adsorbed by exhaustion method	>1.7* [*E. coli*]	n.a.	[49]
	3 [20 $\mu g\ g^{-1}$]	Colloid NPs padded on fabric	99.9 [*S. aureus*] 99.9 [*K. pneumoniae*]	99.9 (10)	[25]
	3–20 [336 $\mu g\ g^{-1}$]	Colloid NPs/PEG adsorbed by exhaustion method	10.5* [*E. coli*] 6.7* [*S. aureus*]	1* [*E. coli*] (50) 1.8* [*S. aureus*] (50)	[133]
	3–20 [336 $\mu g\ g^{-1}$]	Colloid NPs adsorbed by exhaustion method	2–3* [*E. coli*]	n.a.	[39]
	3–20 [894 $\mu g\ g^{-1}$]	Colloid NPs adsorbed by exhaustion method	5–6* [*E. coli*]	n.a.	[39]
	3–8 [108 $\mu g\ g^{-1}$]	Biosynthesized NPs padded on fabric	96 [*E. coli*] 98 [*S. aureus*]	55 [*E. coli*] (20) 59 [*S. aureus*] (20)	[24]
	5	Dodecanethiol-capped NPs in silica sol	40 [*E. coli*]	n.a.	[42]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
	6–8 [100 $\mu g\,g^{-1}$]	Pad-dry-curePad-dry-cure method	96 [*E. coli*] 98.3 [*S. aureus*]	59 [*E. coli*] (20) 62 [*S. aureus*] (20)	[48]
	6–8 [50 $\mu g\,g^{-1}$]	Pad-dry-cure method	96 [*E. coli*] 96.4 [*S. aureus*]	56.6 [*E. coli*] (20) 60.9 [*S. aureus*] (20)	[48]
	7–11 [758 $\mu g\,g^{-1}$]	Microwave synthetized colloid NPs padded on fabric	99.9 [*E. coli*] 99.9 [*S. aureus*]	37 [*E. coli*] (15) 26 [*S. aureus*] (15)	[134]
	8	Pad-dry-cure method	99.9 [*E. coli*] 99.9 [*S. aureus*]	99.9 [*E. coli*] (30) 99.9 [*S. aureus*] (20)	[135]
	10	NPs with dendrimers in Pad-dry-cure method	95 [*E. coli*] 95 [*S. aureus*]	n.a.	[50]
	10 [50 $\mu g\,g^{-1}$]	Colloid NPs impregnated on fabric	99.9 [*E. coli*]99.9 [*S. aureus*] 99.9 [*C. albicans*]	99.9 (5)	[136]
	10–20	Colloid NPs impregnated by US	63.6 [*B. linens*] 62.7 [*S. epidermidis*]	n.a.	[137]
	10–110 [8.2 $\mu g\,g^{-1}$]	Spherical AgNPs deposition by US	99.9 [*S. aureus*] 99.9 [*E. coli*]	0 [*S. aureus*] (5) 0 [*E. coli*] (5)	[34]
	11	NPs grafted with HBP-NH_2	99.4 [*S. aureus*] 99.4 [*E. coli*]	96.7 [*S. aureus*] (50) 96.5 [*E. coli*] (50)	[138]
	15–30	NPs adsorbed by exhaustion method	20 [*F. oxysporum*] 25 [*A. brassicicola*]	n.a.	[139]
	18 [88 $\mu g\,g^{-1}$]	Colloid NPs with HBP-NH2 impregnated on fabric	99 [*E. coli*] 99.3 [*S. aureus*]	98.8 [*E. coli*] (20) 99 [*S. aureus*] (20)	[45]
	20	Covalent bond of AgNPs polystyrene-block-poly-acrylic acid reverse micelle cores	>20* [*E. coli*] >1* [*S. aureus*]	0* [*E. coli*] (5) >1* [*S. aureus*] (20)	[140]
	20 [2 wt.%]		>1.5* [*E. coli*]	n.a.	[141]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
		Colloid NPs impregnated on fabric			
	20–110 [14.1 $\mu g\,g^{-1}$]	Disc AgNPs deposition by US	99.9 [*S. aureus*] 99.9 [*E. coli*]	0 [*S. aureus*] (5) 0 [*E. coli*] (5)	[34]
	20–60 [1500 $\mu g\,g^{-1}$]	UV-assisted Pad-dry-cure method	>1* [*E. coli*] >1.5* [*S. aureus*]	<1* [*E. coli*] (10) >1.5* [*S. aureus*] (10)	[58]
	30 [54 $\mu g\,g^{-1}$]	Colloid NPs adsorbed by exhaustion in CF_4-plasma treated fabric	77 [*P. aeruginosa*] 68 [*E. faecalis*]	n.a.	[60]
	30–50	Colloid NPs adsorbed by exhaustion method	99.9 [*E. coli*] 99.8 [S. epidermis]	93.3 [*E. coli*] (20) 90.8 [*S. epidermis*] (20)	[142]
	30–200 [140 $\mu g\,g^{-1}$]	Silica/AgNPs Pad-dry-cure method	100 [*E. coli*] 100 [*S. aureus*] 100 [*A. niger*]	n.a.	[41]
	32–64 [12.8 $\mu g\,g^{-1}$]	Prism AgNPs deposition by US	99.9 [*S. aureus*] 99.9 [*E. coli*]	12.5 [*S. aureus*] (5) 49.9 [*E. coli*] (5)	[34]
	35–80 [0.5 wt.%]	Colloid NPs deposition by UV	5* [*E. coli*] 4* [*S. aureus*] 5* [*C. albicans*] 3* [*P. p43*]	n.a.	[143]
	41 [5300 $\mu g\,g^{-1}$]	Ethanolic solution of $AgNO_3$ and butylamine impregnated on fabric	98 [*E. coli*] 95 [*S. aureus*]	n.a.	[43]
	50 [30 wt.%]	Gas-phase reaction between phosphine and copper sulphate and $AgNO_3$	100 [*S. aureus*]	100 [*S. aureus*] (10)	[144]
	50 [9.4 $\mu g\,g^{-1}$]	Polygonal AgNPs deposition by US	99.9 [*S. aureus*] 99.9 [*E. coli*]	25 [*S. aureus*] (5) 0 [*E. coli*] (5)	[34]
	60 [100 $\mu g\,g^{-1}$]	NPs and BTCA impregnated on fabric	100 [*E. coli*] 100 [*S. aureus*]	96 [*E. coli*] 92 [*S. aureus*] (30)	[46]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
	75	Microfibers containing NPs by UV irradiation	80 [*E. coli*]	n.a.	[57]
	80 [26 $\mu g\ g^{-1}$]	Colloid NPs adsorbed by exhaustion in CF_4-plasma treated fabric	<60 [*P. aeruginosa*] <60 [*E. faecalis*]	n.a.	[60]
	80 [6 wt.%]	Synthesis and deposition of NPs using US irradiation	100 [*E. coli*] 100 [*S. aureus*]	n.a.	[54]
	180 [4 wt.%]	Colloid NPs padded on fabric	1.66* [*S. aureus*]	4.2* (5)	[23]
	200 [350 $\mu g\ g^{-1}$]	Colloid NPs impregnated on fabric	99 [*E. coli*] 100 [*S. aureus*] 100 [*P. aeruginosa*]	n.a.	[145]
	200 [452 $\mu g\ g^{-1}$]	Colloid NPs padded on fabric	99.9 [*E. coli*] 99.8 [*S. aureus*]	n.a.	[146]
	>200 [16.8 $\mu g\ g^{-1}$]	Hierarchical AgNPs deposition by US	99.9 [*S. aureus*] 99.9 [*E. coli*]	91.3 [*S. aureus*] (5) 99.9 [*E. coli*] (5)	[34]
	257 [34.5 wt.%]	NPs impregnated on fabric	99.9 [*E. coli*]	199.9 [*E. coli*] (10)	[147]
[Ag/Chitosan]	40 [1 wt.%]	Pad-dry-cure method	31* [*E. coli*] 26* [*S. aureus*]	15* [*E. coli*] (20) 17* [*S. aureus*] (20)	[148]
	50–175	Colloid NPs impregnated on fabric	3* [*E. coli*]	n.a.	[52]
[Ag/Chitosan/TiO_2]	5000 [7 wt.%]	Pad-dry-cure method	98 [*E. coli*] 100 [*S. aureus*]	n.a.	[149]
[Chitosan]	5–180 [0.5 wt.%]	Colloid NPs impregnated on fabric	99.9 [*E. coli*] 99.9 [*S. aureus*]	65 [*E. coli*] (20) 78 [*S. aureus*] (20)	[124]
	40	Colloid NPs impregnated on fabric by US	5 [*E. coli*] 25 [*E. faecalis*]	n.a.	[123]
	350 [0.8 wt.%]	NPs grafted with GPTMS	80 [*E. coli*] 80 [*M. lutues*]	n.a.	[125]
[Chitosan/Alginate]	35			95 [*B. cereus*] (30) 87 [*E. coli*] (30) 98 [*P.*	[150]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
		Pad-dry-cure method of NPs loaded with leaf extract	100 [*B. cereus*] 98 [*E. coli*] 100 [*P. aeruginosa*] 100 [*S. aureus*]	*aeruginosa*] (30) 98 [*S. aureus*] (30)	
[CuO]	10 [5 wt.%]	Colloid NPs impregnated on fabric by US	99.9 [*E. coli*] 99.9 [*S. aureus*]	n.a.	[56]
	10–20 [1.5 wt.%]	Colloid NPs impregnated on fabric by US	99.8 [*E. coli*]	n.a.	[55]
	15 [1.4 wt.%]	Colloid NPs impregnated on fabric by US	99.9 [*E. coli*] 99.9 [*S. aureus*]	n.a.	[91]
	40–60 [0.2 wt.%]	Pad-dry-cure method	93.7 [*E. coli*] 95 [*S. aureus*]	48 [*E. coli*] (15) 45 [*S. aureus*] (15)	[151]
	50	Microencapsulated NPs adsorbed by exhaustion method	92.71 [*E. coli*] 100 [*S. aureus*]	86 [*E. coli*] (10) 92 [*S. aureus*] (10)	[38]
	60–75 [2 wt.%]	Pad-dry-cure method	86.5 [*E. coli*] 94.2 [*S. aureus*]	9.8 [*E. coli*] (20) 12 [*S. aureus*] (20)	[92]
	60–80 [0.7 wt.%]	Colloid NPs impregnated on fabric by US	73 [*E. coli*] 66 [*S. aureus*] 72 [*MRSA*] 50 [*A. baumannii*] 74 [*P. aeruginosa*]	5 [*E. coli*] (65) 46 [*S. aureus*] (65)	[95]
	100–150	Colloid NPs coated by pad-dry-cure method	80 [*E. coli*] 99 [*S. aureus*] 98 [*A.niger*]	n.a.	[93]
	200–400 [0.74 wt.%]	Colloid NPs impregnated on fabric by US	38 [*E. coli*] 38 [*S. aureus*] 52 [*K. pneumonia*] 52 [*MRSA*] 1 [*A. baumannii*] 15 [*P. aeruginosa*]	n.a.	[152]
[TiO_2]	7 [2 wt.%]	Pad-dry-cure method	72.9 [*S. aureus*] 74.5 [*K. pneumonia*]	29.9 [*S. aureus*] (20) 30.5 [*K. pneumonia*] (20)	[153]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
	10–15 [6 wt.%]	Colloid anatase NPs impregnated on fabric by US	25 [*E. coli*] 94.4 [*S. aureus*] 59.5 [*C. albicans*]	n.a.	[154]
	10–15 [8 wt.%]	Colloid rutile NPs impregnated on fabric by US	6.9 [*E. coli*] 72.4 [*S. aureus*] 40.3 [*C. albicans*]	n.a.	[154]
	10–15 [6 wt.%]	Colloid anatase NPs impregnated on fabric by US + UV	29.2 [*E. coli*] 99.9 [*S. aureus*] 70.1 [*C. albicans*]	n.a.	[154]
	10–15 [8 wt.%]	Colloid rutile NPs impregnated on fabric by US + UV	31.9 [*E. coli*] 99.3 [*S. aureus*] 62.4 [*C. albicans*]	n.a.	[154]
	12m [2 wt.%]	Pad-dry-cure method	75.8 [*S. aureus*] 77.6 [*K. pneumonia*]	34.7 [*S. aureus*] (20) 34.7 [*K. pneumonia*] (20)	[153]
	13–20 [1 μg g^{-1}]	Pad-dry-cure method	94 [*E. coli*] 99 [*S. aureus*]	83 [*E. coli*] (10) 86 [*S. aureus*] (10)	[76]
	<50 [5 wt.%]	Colloid NPs impregnated on fabric	98 [*S. aureus*] 98 [*K. pneumoniae*]	n.a.	[75]
	70–390 [0.5 wt.%]	Apatite-coated NPs by pad-dry-cure method	5.5 [*E. coli*] 13.4 [*S. aureus*] 24.2 [*M. luteus*]	n.a.	[74]
[ZnO]	10–20 [0.8 wt.%]	Colloid NPs impregnated on fabric by US	17 [*E. coli*]	n.a.	[55]
	20–100	Colloid NPs impregnated on fabric by US	100 [*S. aureus*]	n.a.	[155]
	25	ZnO nanoparticle incorporated PS-b-PAA coating	>1* [*S. aureus*] >1* [*E. coli*]	n.a.	[156]
	30–40 [0.66 wt.%]	Roll to roll US coating on enzyme pre-treated fabric	36 [*S. aureus*] 35 [*P. aeruginosa*] 25 [*A.*	15 [*S. aureus*] (10) 30 [*P. aeruginosa*] (10) 12 [*A. baumannii*] (10) 30	[157]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
			baumannii] 70 [*E. coli*] 31 [MRSA]	[*E. coli*] (10) 32 [MRSA] (10)	
	30–60 [10 wt.%]	Pad-dry-cure method	99 [*E. coli*] 98 [*M. luteus*]	n.a.	[158]
	30 [0.75 wt.%]	Colloid NPs deposition by US	99.9 [*E. coli*] 99.9 [*S. aureus*]	n.a.	[83]
	37 [20 wt.%]	Pad-dry-cure method	100 [*E. coli*] 100 [*S. aureus*]	n.a.	[159]
	38 [0.2 wt.%]	Pad-dry-cure method	91.8 [*S. aureus*] 15 [*K. pneumoniae*]	n.a.	[84]
	38 [0.6 wt.%]	Pad-dry-cure method	96.8 [*S. aureus*] 99.9 [*K. pneumoniae*]	n.a.	[84]
	38 [1 wt.%]	Pad-dry-cure method	99.9 [*S. aureus*] 99.9 [*K. pneumoniae*]	n.a.	[84]
	<50	Colloid NPs impregnated on fabric	97 [*S. aureus*] 98 [*K. pneumoniae*]	n.a.	[75]
	60–70 [0.75 wt.%]	Roll to roll US coating on fabric	36 [*S. aureus*] 18 [*P. aeruginosa*] 11 [*A. baumannii*] 65 [*E. coli*] 38 [*MRSA*]	15 [*S. aureus*] (10) 12 [*P. aeruginosa*] (10) 0 [*A. baumannii*] (10) 5 [*E. coli*] (10) 10 [*MRSA*] (10)	[157]
	<100	Layer-by-layer deposition of NPs	1.3* [*S. aureus*]	0 [*S. aureus*] (20)	[85]
	80–150 [4.3 wt.%]	Nanorods and chalcone solution padded on fabric	99.9 [*E. coli*] 99.6 [*S. aureus*] 99.9 [*P. aeruginosa*]	n.a.	[36]
	200 [2 wt.%]	Pad-dry-cure method	80 [*E. coli*] 99.6 [*S. aureus*]	75 [*E. coli*] (5) 98 [*S. aureus*] (5)	[160]
[ZnO/Chitosan]	15–60 [0.3 wt.%]	Colloid NPs impregnated on fabric by US	99.9 [*E. coli*] 98.5 [*S. aureus*]	85 [*E. coli*] (10) 70 [*S. aureus*] (10)	[161]
	28–100 [6 wt.%]	Pad-dry-cure method		n.a.	[89]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
			22* [*E. coli*] 25* [*S. aureus*]		
	30	Pad-dry-cure method	100 [*E. coli*] 100 [*M. luteus*]	n.a.	[162]
	60 [0.5 wt.%]	Colloid NPs impregnated on fabric by US	40 [*E. coli*] 48 [*E. faecalis*]	n.a.	[123]
[ZrO]	2–5 [2.41 wt.%]	Colloid NPs synthetized and adsorbed on fabric	98 [*S. typhimurium*] 95 [*S. aureus*]	92 [*S. typhimurium*] (30) 90 [*S. aureus*] (30)	[98]
Cotton/polyester					
[Ag]	30–200 [59 $\mu g\, g^{-1}$]	Silica/AgNPs Pad-dry-cure method (65/35)	100 [*E. coli*] 100 [*S. aureus*] 100 [*A. niger*]	n.a.	[41]
	100 [1000 $\mu g\, g^{-1}$]	Colloid NPs impregnated on fabric (60/40)	100 [*S. aureus*]	100 [*S. aureus*] (4)	[163]
[TiO_2]	<50 [5 wt.%]	Colloid NPs impregnated on fabric (55/45)	98 [*S. aureus*] 99 [*K. pneumoniae*]	n.a.	[75]
[ZnO]	30	Pad-dry-cure method	100 [*E. coli*] 100 [*M. luteus*]	n.a.	[162]
	30–60 [10 wt.%]	Pad-dry-cure method (65/35)	98 [*E. coli*] 99 [*M. luteus*]	n.a.	[158]
	<50	Colloid NPs impregnated on fabric (55/45)	98 [*S. aureus*] 99 [*K. pneumoniae*]	n.a.	[75]
Polyacrylonitrile					
[Chitosan]	1000 [15 wt.%]	Electrospinned nanofibers containing Chitosan	100 [*E. coli*] 100 [*S. aureus*] 99.8 [*P. aeruginosa*] 100 [*M. luteus*]	n.a.	[122]
Polyamide					
[Ag]	8	Electrospinned nanofibers containing NPs	99.9 [*E. coli*]	n.a.	[105]
	10 [4.46 $\mu g\, g^{-1}$]		99.9 [*C. albicans*]	64.7 [*C. albicans*] (5)	[70]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
		Colloid NPs impregnated on corona-plasma treated fabric			
	10 [4.46 μg g^{-1}]	Colloid NPs impregnated on corona-plasma treated fabric	99.9 [*E. coli*] 99.9 [*S. aureus*]	83.2 [*E. coli*] (5) 85.3 [*S. aureus*] (5)	[63]
	10 [4.46 μg g^{-1}]	Colloid NPs impregnated on corona-plasma treated dyed fabric	98.6 [*C. albicans*]	n.a.	[64]
	10 [4.46 μg g^{-1}]	Colloid NPs impregnated on corona-plasma treated dyed fabric	99.9 [*E. coli*] 99.9 [*S. aureus*]	n.a.	[164]
	10–20 [0.025 wt.%]	Thermal reduction of silver acetate during melt processing of PA	80.6 [*E. coli*]	n.a.	[101]
	10–20 [0.06 wt.%]	Thermal reduction of silver acetate during melt processing of PA	99.9 [*E. coli*]	n.a.	[101]
	30–200 [31 μg g^{-1}]	Silica/AgNPs Pad-dry-cure method	100 [*E. coli*] 100 [*S. aureus*] 100 [*A. niger*]	n.a.	[41]
	50–100 [1 wt.%]	Colloid NPs impregnated on fabric by US	99 [*P. aeruginosa*] 99 [*S. aureus*]	n.a.	[66]
	70	Colloid NPs impregnated on fabric	5–6* [*E. coli*]	n.a.	[165]
	80	Colloid NPs impregnated on dyed fabric	100 [*E. coli*] 100 [*S. aureus*] 100 [*P. aeruginosa*]	0 [*E. coli*] 0 [*S. aureus*] 0 [*P. aeruginosa*]	[166]
	<100	Layer-by-layer deposition of NPs	53 [*S. aureus*]	n.a.	[167]
[CuO]	85 [8.5 wt.%]		>1* [*S. aureus*]	n.a.	[97]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
		In situ produced NPs grafted with CTAB			
[TiO_2]	21 [1 wt.%]	Electrospinned nanofibers containing NPs	99 [*E. coli*]	n.a.	[73]
[ZnO]	<100 [5 wt.%]	Sheath-core fibers prepared by melt-spinning method	100 [*S. aureus*] 100 [*K. pneumoniae*]	n.a.	[86]
	<100 [1 wt.%]	Sheath-core fibers prepared by melt-spinning method	95 [*S. aureus*] 68 [*K. pneumoniae*]	n.a.	[86]
Poly(ε-caprolactone)					
[Ag-$Zr(HPO_4)_2$]	63.7 [1 wt.%]	Electrospinned nanofibers containing NPs	98.4 [*E. coli*] 99.3 [*S. aureus*]	n.a.	[106]
Polyester					
[Ag]	2–5	Colloid NPs padded on fabric	99.7 [*S. aureus*] 99.8 [*K. pneumoniae*]	15.3 [*K. pneumoniae*] (20) 84.3 [*S. aureus*] (20)	[132]
	2–6	Silver–poly(acrylate) NPs clusters impregnated on fabric	>1* [*S. aureus*] >1* [*S. epidermidis*] >1* [*P. aeruginosa*] >1* [*C. albicans*]	n.a.	[44]
	3 [10 $\mu g\, g^{-1}$]	Colloid NPs padded on fabric	99.9 [*S. aureus*] 99.9 [*K. pneumoniae*]	n.a.	[25]
	10	NPs with dendrimers in Pad-dry-cure method	95 [*E. coli*] 60 [*S. aureus*]	n.a.	[50]
	10 [8.61 $\mu g\, g^{-1}$]	Colloid NPs impregnated on corona-plasma treated fabric	99.1 [*C. albicans*]	96.7 [*C. albicans*] (5)	[70]
	10 [8.61 $\mu g\, g^{-1}$]				[63]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
		Colloid NPs impregnated on corona-plasma treated fabric	99.9 [*E. coli*] 99.8 [*S. aureus*]	99.9 [*E. coli*] (5) 99.6 [*S. aureus*] (5)	
	10 [8.61 $\mu g\ g^{-1}$]	Colloid NPs impregnated on corona-plasma treated dyed fabric	99.9 [*C. albicans*]	n.a.	[64]
	10 [8.61 $\mu g\ g^{-1}$]	Colloid NPs impregnated on corona-plasma treated dyed fabric	99.9 [*E. coli*] 99.9 [*S. aureus*]	n.a.	[164]
	11 [1 wt.%]	NPs plasma grafted with acrylic acid	99.9 [*E. coli*] 99.9 [*S. aureus*]	n.a.	[168]
	<20 [10 $\mu g\ g^{-1}$]	Colloid NPs impregnated on fabric	100 [*S. aureus*] 42 [*K. pneumoniae*]	n.a.	[169]
	<20 [100 $\mu g\ g^{-1}$]	Colloid NPs impregnated on fabric	100 [*S. aureus*] 100 [*K. pneumoniae*]	n.a.	[169]
	30–200 [8.9 $\mu g\ g^{-1}$]	Silica/AgNPs Pad-dry-cure method	100 [*E. coli*] 100 [*S. aureus*] 100 [*A. niger*]	n.a.	[41]
	40–70 [155 $\mu g\ g^{-1}$]	Colloid NPs impregnated on RF-plasma treated fabric	99.9 [*E. coli*] 99.9 [*S. aureus*]	99.9 [*E. coli*] 5) 99.9 [*S. aureus*] 5)	[61]
	80 [93 $\mu g\ g^{-1}$]	Colloid NPs impregnated on corona-plasma treated fabric	19 [*E. coli*] 67 [*S. aureus*] 74 [*S. faecalis*] 6 [*P. aeruginosa*]	n.a.	[62]
[Ag/Chitosan]	166 [0.2 wt.%]	Colloid NPs impregnated on PVP treated fabric	100 [*S. aureus*]	n.a.	[53]
[Chitosan]	5–180 [0.5 wt.%]	Colloid NPs impregnated on fabric	90 [*E. coli*] 99.9 [*S. aureus*]	50 [*E. coli*] (20) 75 [*S. aureus*] (20)	[124]
	115 [0.2 wt.%]	Colloid NPs impregnated on PVP treated fabric	90 [*S. aureus*]	n.a.	[53]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
[Fe_3O_4]	30–40	In situ synthesis of NPs	100 [*S. aureus*]	n.a.	[100]
[α-Fe_2O_3]	40–50	In situ synthesis of NPs	80.5 [*S. aureus*]	n.a.	[100]
[SiO_2/Ag/CuO]	500/30/17 [2.5 wt.%]	Top-coating with Pericoat PU 340 NEW paste containing NPs	100 [*E. coli*] 99.8 [*S. aureus*] 99.9 [*K. pneumoniae*] 100 [*C. albicans*] 99.4 [*A. niger*] 96.5 [*T. mentagraphytes*]	99.9 [*E. coli*] (20) 99.8 [*S. aureus*] (20) 99.9 [*K. pneumoniae*] (20) 100 [*C. albicans*] (20) 56.4 [*A. niger*] (20) 92.1 [*T. mentagraphytes*] (20)	[170]
[TiO_2]	6 [2.1 wt.%]	Alginates and colloid NPs impregnated on fabric	99.9 [*E. coli*]	99.8 (5)	[171]
Polyethylene [Ag/Chitosan]	5 [1.1 wt.%]	Electrospinned nanofibers containing NPs	99.9 [*E. coli*]	n.a.	[107]
Polyethylene/ Chitosan [Ag]	12–18 [1.3 wt.%]	Electrospinned nanofibers containing NPs	15* [*E. coli*] 20* [*S. aureus*] 18* [*P. aeruginosa*] 12* [*C. albicans*]	n.a.	[172]
Polyethylene/ polypropylene [Ag]	<10 [12 $\mu g\, g^{-1}$]	Colloid NPs padded on non-woven fabric	99.8 [*S. aureus*] 99.9 [*K. pneumoniae*]	n.a.	[173]
Poly(l-lactide) [Ag]	30 [32 wt.%]	Electrospinned nanofibers containing NPs	94.2 [*E. coli*] 98.5 [*S. aureus*]	n.a.	[108]
	35 [5 wt.%]	Electrospinned nanofibers containing NPs	5* [*E. coli*] 5* [*S. aureus*]	n.a.	[109]
Polypropylene [Ag]	15 [0.3 wt.%]			n.a.	[103]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
		Sheath-core fibers prepared by melt-spinning method	99.9 [*S. aureus*] 99.9 [*K. pneumoniae*]		
	30 [0.1 wt.%]	Twin-screw mixer extrusion	99.9 [*S. aureus*]	n.a.	[102]
[TiO_2/Ag]	60–100 [0.2 wt.%]	Sheath-core fibers prepared by melt-spinning method	99.2 [*S. aureus*]	n.a.	[78]
Poly(vinyl alcohol)					
[Ag]	6 [0.1 wt.%]	Electrospinned nanofibers containing NPs	99.9 [*S. aureus*] 99.9 [*K. pneumoniae*]	n.a.	[110]
[Ag/Chitosan]	2–10 [1 wt.%]	Electrospinned nanofibers containing NPs	99.9 [*E. coli*]	n.a.	[111]
	20 [0.6 wt.%]	Electrospinned PVA nanofibers containing NPs	100 [*E. coli*]	n.a.	[51]
[TiO_2/Ag/Chitosan]	100 [0.04 wt.%]	Electrospinned nanofibers containing NPs	99 [*E. coli*] 98 [*S. aureus*]	n.a.	[79]
Poly(vinyl alcohol)/Silk					
[Ag]	3.8	Electrospinned nanofibers containing NPs	>1* [*E. coli*] >1* [*S. aureus*]	n.a.	[112]
Silk					
[Ag]	4.3 [116.5 $\mu g\, g^{-1}$]	Colloid NPs and PNP impregnated on fabric	99.5 [*E. coli*] 99.9 [*S. aureus*]	98.9 [*E. coli*] (30) 99.4 [*S. aureus*] (30)	[174]
	5–50 [2.3 wt.%]	Colloid NPs impregnated on fabric	99.9 [*S. aureus*]	n.a.	[175]
	15–30	Colloid NPs adsorbed by exhaustion method	45 [*F. oxysporum*] 50 [*A. brassicola*]	n.a.	[139]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
	10 [40 $\mu g\,g^{-1}$]	Colloid NPs applied by exhaustion	100 [*S. aureus*]	84 [*S. aureus*] (10)	[176]
	<10	UV-assisted in situ synthesis of AgNPs	1.59* [*E. coli*] 1.84* [*S. aureus*]	n.a.	[65]
	<10 [50 $\mu g\,g^{-1}$]	Colloid NPs applied by exhaustion in US bath	94 [*E. coli*] 100 [*S. aureus*]	n.a.	[177]
	11.5 [13 $\mu g\,g^{-1}$]	Colloid NPs impregnated on fabric	>1* [*M. lysodeikticus*] >1* [*E. coli*] >1* [*S. aureus*]	n.a.	[178]
	<20	Synthesis of NPs on silk fibers via γ-ray irradiation	96 [*S. aureus*]	85 [*S. aureus*] (10)	[179]
	30–200 [170 $\mu g\,g^{-1}$]	Silica/AgNPs Pad-dry-cure method	100 [*E. coli*] 100 [*S. aureus*] 100 [*A. niger*]	n.a.	[41]
	35 [60 $\mu g\,g^{-1}$]	Colloid NPs applied by exhaustion	100 [*S. aureus*]	78 [*S. aureus*] (10)	[176]
	50 [268.6 $\mu g\,g^{-1}$]	Colloid NPs impregnated on fabric	99 [*E. coli*]	98 [*E. coli*] (50)	[180]
	<100 [98.7 $\mu g\,g^{-1}$]	Multi-amidine/silver nitrate sol added by steam method	99.9 [*E. coli*] 99.5 [*S. aureus*]	>97.4 [*E. coli*] (50)	[181]
	<100	Colloid NPs impregnated on fabric	100 [*E. coli*]	n.a.	[182]
	<100	Layer-by-layer deposition of NPs	80 [*S. aureus*]	n.a.	[167]
[Au]	21 [0.21 wt.%]	*In situ* synthesized NPs impregnated on fabric	100 [*E. coli*]	n.a.	[99]
[Chitosan]	20.8 [1 wt.%]	Colloid NPs impregnated on fabric	95 [*S. aureus*]	90 [*S. aureus*] (20)	[183]
[TiO_2]	50 [25 $\mu g\,g^{-1}$]			n.a.	[184]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
		Colloid NPs and PUA applied by dyeing	19* [*E. coli*] 23* [*S. aureus*]		
[TiO_2/Ag]	20/5 [1 wt.%]	DHPBA modified NPs padded on fabric	>1* [*E. coli*] >1* [*S. aureus*] >1* [*P. aeruginosa*]	n.a.	[80]
Viscose					
[Ag]	2–9 [4 wt.%]	AgNPs SiO_2 Sol-gel coating	53 [*A. niger*] 41 [*B. subtilis*] 88 [*P. putida*]	n.a.	[185]
	30–200 [230 $\mu g\, g^{-1}$]	Silica/AgNPs Pad-dry-cure method	100 [*E. coli*] 100 [*S. aureus*] 100 [*A. niger*]	n.a.	[41]
[Chitosan]	5–180 [0.5 wt.%]	Colloid NPs impregnated on fabric	99.9 [*E. coli*] 99.9 [*S. aureus*]	55 [*E. coli*] (20) 76 [*S. aureus*] (20)	[124]
Wool					
[Ag]	1–7	Photocured Carboxymethyl Starch NPs	24* [*S. aureus*] 22* [*E. coli*]	3* [*S. aureus*] (15) 1* [*E. coli*] (15)	[127]
	2–6	Silver–poly(acrylate) NPs clusters impregnated on fabric	>1* [*C. albicans*]	n.a.	[44]
	4.2 [5 $\mu g\, g^{-1}$]	Colloid silver NPs impregnated by pad-dry-cure	99.9 [*S. aureus*] 99.7 [*K. pneumoniae*]	n.a.	[186]
	15 × 6	Nanodisc colloid NPs impregnated on fabric	98.5 [*E. coli*]	n.a.	[35]
	22 × 14	Nanodisc colloid NPs impregnated on fabric	93.8 [*E. coli*]	n.a.	[35]
	45–60	Colloid NPs adsorbed by exhaustion method	99.9 [*E. coli*] 98.9 [*S. epidermis*]	92 [*E. coli*] (20) 91.7 [*S. epidermis*] (20)	[142]
	48 × 5	Nanoprism colloid NPs impregnated on fabric	77 [*E. coli*]	n.a.	[35]
				n.a.	[41]

Table 1. (Continued.)

Textile fabric and fiber [Nanomaterial]	NPs average size (nm) [Concentration]	Method	% Bacterial reduction [Strain]	% Bacterial reduction after washing [Strain] (Washing cycles)	Reference
	30–200 [310 $\mu g\ g^{-1}$]	Silica/AgNPs Pad-dry-cure method	18 [*E. coli*] 56 [*S. aureus*] 50 [*A.niger*]		
[SiO_2/Ag]	34.6	Colloid NPs impregnated on fabric	71 [*E. coli*]	n.a.	[187]
	60 [4.3 wt.%]	Colloid NPs impregnated on RF-plasma treated fabric	85 [*E. coli*] 95 [*S. aureus*]	70 [*E. coli*] (20) 73 [*S. aureus*] (20)	[71]
[TiO_2/Ag]	21 [1 wt.%]	TiO_2 NPs in silver nitrate solution impregnated on fabric	100 [*E. coli*] 100 [*S. aureus*]	n.a.	[81]
Wool/polyester					
[Ag]	30–200 [250 $\mu g\ g^{-1}$]	Silica/AgNPs Pad-dry-cure method (45/55)	100 [*E. coli*] 100 [*S. aureus*] 0 [*A.niger*]	n.a.	[41]
[TiO_2]	21 [0.25 wt.%]	Colloid NPs crosslinked with BTCA on fabric (45/55)	99 [*E. coli*]	n.a.	[188]
	21 [0.75 wt.%]	Colloid NPs crosslinked with BTCA on fabric (45/55)	100 [*E. coli*]	n.a.	[188]

*Inhibition zone in mm; n.a. Not available.

substrates without affecting their macro-scale properties, such as breathability or hand feel [18]. Nanoparticles as antimicrobial agents have increasingly been used in textile research due to their unique physico-chemical properties and biological activity, which may differ significantly from ion and bulk materials [21]. However, nanoparticles (NPs) in humans may affect normal cellular proliferation and protein functions primarily due to their metallic nature, and the generation of reactive oxygen species may initiate pro-inflammatory and toxic activities [22]. The first chapter of the manuscript is dedicated to AgNPs because most of the research in this field was conducted using this metal. For the same reason, because the majority of the information regarding the effect of NP morphology and deposition methods was only studied on silver, three subchapters (NP morphology, chemical deposition methods and physical deposition methods) were added. The first subchapter is dedicated to the importance of size, shape, composition, crystallinity and structure of NPs on their antimicrobial activity. The second is dedicated to chemical deposition methods, with special emphasis on the new routes for the deposition of NPs based on environmental benign natural polymers. The third subchapter is about physical deposition methods, focusing on plasma technology. The following chapters report on research using other antimicrobial metal and metal oxide NPs and nanofibers. The last chapter reports on the recent use of natural polymer NPs, especially chitosan, in the antimicrobial finishing of textiles due to the environmental and toxicological concerns regarding the use of heavy metals for the production of NPs. Thus, due to the increasing dichotomy between environmental and health concerns and the potential benefits of using NPs as finishing agents, this review offer the opportunity to update and critically discuss the latest advances and applications for the textile industry.

Nanosilver

As we can see in table 1, several methods have been used for surface nanomodifications of textiles, but most of the research has been performed using nanosilver immobilized on cotton, polyester, polyamide, silk and wool fabrics by conventional dip- or pad-dry methods [23–25]. The term 'nanosilver' is conventionally attributed to silver metals, but a fraction of the silver salts could also fall under the NP definition according to the International Standard Organisation (ISO), which defines an NP as having a maximum diameter of 100 nm in at least three dimensions. However, all particles with a diameter between 100 to 1000 nm were also assumed to contain NPs, unless there was concrete information about the size distribution and the stability of agglomerates. Taking this into account, according to Windler *et al* up to 80% of all silver used in textiles (45 metric tonnes) may be considered in the nanoform [26]. Considerable work has been done in the functionalization of textile materials with AgNPs. Most of the research is focused on the antimicrobial effects of modified textile materials, but no evident conclusions about the binding mechanism between AgNPs and textile fibers has been proposed. Thus, due to the huge amount of AgNP-containing textiles, some concerns have been raised about the release of silver into the environment after repeated washing [27, 28]. Some authors questioning the use of AgNPs of lower than 30 nm in textiles due to the additional effort it requires towards synthesis, stabilization and incorporation when the same results could be obtained simply by immersing the fabric in solutions of $AgNO_3$ [29]. The critics are basing their questions on the difficulties in achieving small particle sizes of narrow size distributions with green processes and on the toxicity concerns of particles with a size between 1 and 10 nm,

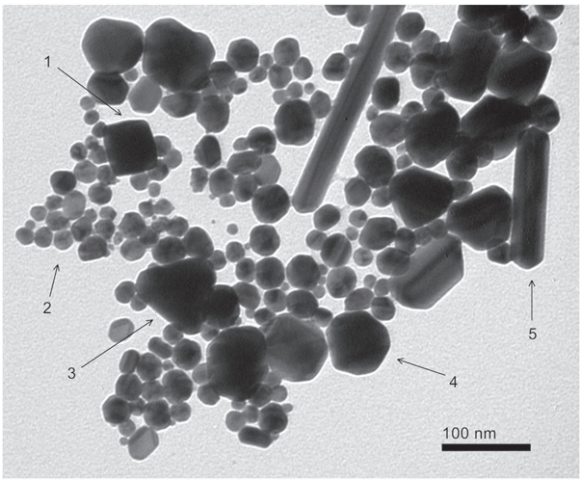

Figure 1. Close-up of the TEM image of AgNPs (X200000 magnification) in different shapes and sizes. (1) Cubic, (2) Spherical ~10 nm, (3) Triangular, (4) Spherical ~60 nm, (5) Rod-like. TEM was performed using a JEOL JEM 1400 TEM (Tokyo, Japan) operating at an acceleration voltage of 120 kV. Nanoparticle sample was applied to glow-discharged carbon-coated copper grids followed by negative staining with a solution of 1% (w/v) uranyl acetate.

which can penetrate human skin [30]. Nowadays, the release of silver is an emerging environmental problem. Therefore, it is expected that further research will be more oriented towards the environmental, nanotechnological and regulatory aspects of the exploitation of textile products with deposited or immobilized NPs [31].

NP morphology

The way in which NPs alter surface properties and impart textile antimicrobial functions is mainly determined by their size, shape, composition, crystallinity and structure [32]. It has been described that AgNPs of between 1 and 10 nm present a greater impact on bacteria than larger particles, and that triangular-shaped NPs display greater biocidal action than rod- or spherical-shaped ones [33]. Recent results of studies on the antimicrobial effect on cotton fabric of different morphologies of AgNPs, such as spherical, polygonal, disk, prism, and hierarchical assemblies (figure 1), confirmed that non-spherical morphologies, such as polygonal-, prism- and hierarchical-like shapes, in comparison with spherical and disc morphologies exhibited a stronger growth-inhibitory effect against Gram-negative and Gram-positive bacteria. In addition, among various tested morphologies, the hierarchical-like morphology showed very good antimicrobial activity after five washing cycles [34]. On the other hand, in a few other works available in the literature using nano-rods, -discs and -prisms in textiles, moderate biocide activity is shown when compared with conventional spherical-shaped NPs [35, 36]. Supposedly, the significantly larger surface area of the NPs allows higher contact with bacteria, enhancing their bactericidal activity. However, other factors, such as dielectric and quantum confinement effects, could be responsible for the different properties of metal or metal-oxide NPs with respect to bulk materials [37].

Chemical deposition methods

Most of the methods used in AgNP production are based on reactions in the liquid medium and often require environmentally hazardous surfactants, reducing agents and templates for the synthesis of AgNPs [38, 39]. Several studies reporting on silica–silver core–shell NPs mainly review the chemical synthesis processes and their characterization. Silica is a class of very important core materials for immobilizing NPs on its surface due to its high chemical and thermal stability, chemical inertness, large surface areas, and good compatibility with other materials. Nischala *et al* synthesized extremely small (1–2 nm) AgNPs attached on silica core particles with an average size of 270 nm using a simple one-pot chemical method. The silver containing silica core–shell particles immobilized on cotton did not leach out from the fabric and showed excellent antibacterial activity even after 10 washing cycles [40]. Novel fiber–silica–Ag composites with biocidal activity were also successfully produced by chemically modifying cotton, wool, silk, polyester and polyamide fabrics. The results show that the chemical and morphological structures of the fibers directly influenced their absorptivity and affinity for the AgNPs. On the other hand, a chemical strong-binding of Ag to the fibers seems to significantly reduce the effectiveness of the antimicrobial activity of the AgNPs [41]. Other methods include the reduction of silver ions by ethanol or isopropanol or the coating with acrylates or cross-linkable polysiloxanes to stabilize NP dispersion onto the fabric [32, 42–46]. The introduction of green chemistry into nanotechnology is one of the most important topics in nanoscience research today. The main purpose is to avoid the environmental toxicity or biological hazards normally associated with the preparation of AgNPs using synthetic reducing agents. To date, new routes for the development of NPs based on environmentally benign natural polymers such as chitosan, hyaluronan, starch, and cyclodextrin have been explored [47]. Hebeish *et al* synthesized small AgNPs using hydroxypropyl starch as both a reducing and stabilizing agent, retaining excellent antibacterial properties even after 20 washing cycles, reflecting the importance of binders in the fixation of AgNPs on the surface of the fabrics [48]. Raghavendra *et al* tested on cellulose fibers several natural carbohydrates such as gum acacia and gaur gum as an effective reducing agent for the green synthesis of AgNPs from $AgNO_3$. The thermal stability and mechanical properties of the cellulosic composites were found to be better than cellulose fibers alone [49]. Mahltig *et al* fabricated hybrid nanomaterials based on dendrimers as polymeric stabilizers for the preparation of AgNPs used as finishing agents to produce antimicrobial textiles. The results confirmed that the antimicrobial effect rises with increases in the dendrimers' generations due to decreasing size of the formed AgNPs. By changing thermal fixation and dendrimers' generations, the strength of the antimicrobial effect can be controlled [50]. Several works involve chemical modification of textile fabrics by natural, biocompatible and biodegradable polysaccharide chitosan followed by incorporating AgNPs into the fabrics. Abdelgawad *et al* produced antibacterial nanofiber mats of PVA loaded with AgNPs enveloped in chitosan after reduction with glucose. The results showed superior properties and synergistic antibacterial effects by combining chitosan with AgNPs [51]. Other authors produced silver-loaded chitosan NPs attached to textiles, which also exhibited excellent antibacterial activity [52, 53].

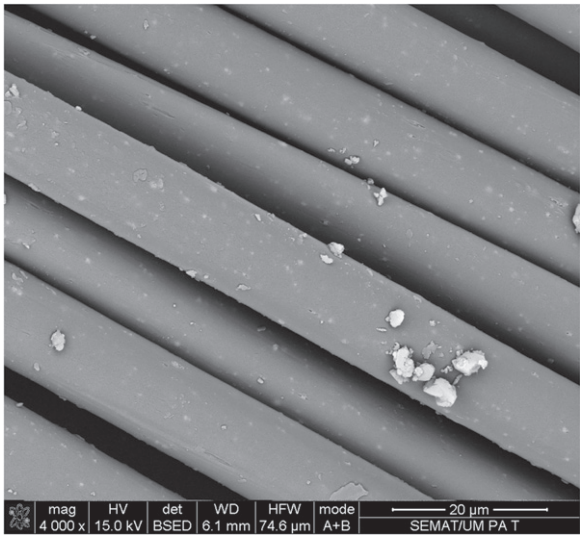

Figure 2. SEM images (X4000 magnification) of antimicrobial AgNP aggregates deposited on DBD plasma pre-treated polyamide 6,6 fibers. Images were carried in a FEG-SEM, NOVA 200 Nano SEM, FEI Company. Secondary and backscattering electron images were performed with an acceleration voltage of 5 kV and 15 kV, respectively. Samples were covered with a film of Au–Pd (80–20 wt.%) in a high-resolution sputter coater, 208 h Cressington Company, coupled to a MTM-20 Cressington High Resolution Thickness Controller.

Physical deposition methods

In the last decade, physical methods such as ultrasound [54–56], UV irradiation [57–59], plasma pre-treatment and ion-beam-assisted deposition [60–64] have been proved to be effective for the deposition, insertion and synthesis of well-dispersed nanophase materials on textiles. Lu *et al* developed a UV-assisted *in situ* synthesis approach to immobilize AgNPs on silk fibers for antibacterial applications. Results show that AgNPs with excellent crystalline structures are efficiently attached on the silk surface in an irradiation time-dependent manner [65]. Sonochemical reactions are capable of enhancing AgNP adhesion to the fabric surface by physical or chemical bonding depending on the nature of the substrate [66]. Pre-treatment of textiles by low-pressure plasmas can also improve loading of AgNPs from colloids (figure 2). Different plasma particles (e.g. electrons, ions, free radicals, photons) provide superficial functionalization and etching of the fiber without deterioration of bulk properties. Plasma is particularly important for the surface activation of hydrophobic synthetic fibers such as polyester and polyamide fabrics because it makes fibers more accessible to water and chemical species [67–69]. However, little research using plasma pre-treatment reports about antimicrobial activity on fabrics. The Serbian group headed by M Radetic reported great stability and uniform AgNPs coatings, as well as high antibacterial activity and laundering durability, using several plasma sources such as low-temperature air radio frequency (RF), dielectric barrier discharge (DBD) and corona discharge in different textile materials [61, 63, 64, 70]. Although RF-powered plasma devices allow easier control of properties and uniformity, this system requires more complex handling and a vacuum system, which can be avoided by using DBD and corona discharges at atmospheric pressure. Other groups working with plasma (including corona and

CF_4-plasma) have obtained similar results on AgNPs deposition, but with lower biocide performance [60, 62, 71].

Nanotitanium dioxide

Nano TiO_2, one of the most powerful photocatalytic materials, possesses high activity, strong oxidizing power and long-term stability [72]. When illuminated under UV light with wavelengths lower than 385 nm, nano TiO_2 electrons are excited from the valence band to the conduction band [73]. The positive hole in the valence band can then react with water or hydroxide ions adsorbed on the surface to produce hydroxyl radicals, and the electron in the conduction band can reduce O_2 to produce superoxide ions. These two highly reactive species are able to decompose a variety of organic materials, including microorganisms [74]. However, there are few reports on the use of TiO_2 nanomaterial for textile applications, and only NPs with a diameter lower than 20 nm have shown effective, but not complete, antimicrobial activity in cotton, polyester, polyamide and wool/polyester fabrics [73–75].

Khurana *et al* observed that cationic as well as non-ionic dispersing agents led to a reduction in size of the TiO_2 NPs produced by sol gel methods, whereas anionic dispersing agents led to an increase in particle size. The TiO_2 NPs so synthesized were successfully applied onto cotton while maintaining their antimicrobial activity for up to 10 washes with the help of a binder [76]. Although there are numerous advantages in utilizing nano TiO_2 in textiles, some drawbacks have also been reported. First, due to its high band gap, semiconductor TiO_2 shows photocatalytic activity under UV rays, which practically limits the use of sunlight or visible light as an irradiation source. Second, the electron-hole recombination rate is too high, resulting in low photocatalytic efficiency. It has been suggested that through adding noble metals to the surface of TiO_2, photocatalytic activity can be increased by extending the light absorption range of TiO_2 from UV to the visible range [77]. Some examples using mixed silver/TiO_2 are found in the existing literature for polypropylene, poly (vinyl alcohol), silk and wool, but they did not apparently show any additional advantages over silver [78–81]. Moreover, the dissipation mechanism of the UV energy is not often considered. A direct application of TiO_2 to products such as paint, textile, plastics and paper can lead to the creation of free radicals with consequent photochemical decomposition of the substrates [74]. Free radicals are also implicated in a number of potential health issues such as skin aging. However, free radical generation can be reduced by over 90% by incorporating a dopant ion within the titanium oxide lattice structure [82].

Nanozinc oxide

ZnO NPs exhibit strong antibacterial activities on a broad spectrum of bacteria on cotton [36, 55, 75, 83–85], polyamide [86] and bamboo fabric [87]. Moreover, excellent multi-functional textiles with good UV protection in addition to very good antibacterial properties against Gram positive and Gram negative bacteria can be obtained using ZnO in combination with synthetic [88] and natural organic polymers such as chitosan [89]. The use of functional polymer matrices such as PMME or PNIPAM as a dispersion medium for ZnO NPs results in improved functional and bonding properties in fabrics (figure 3). Similar to TiO_2, the photocatalytic generation of hydrogen peroxide was suggested to be one of the primary

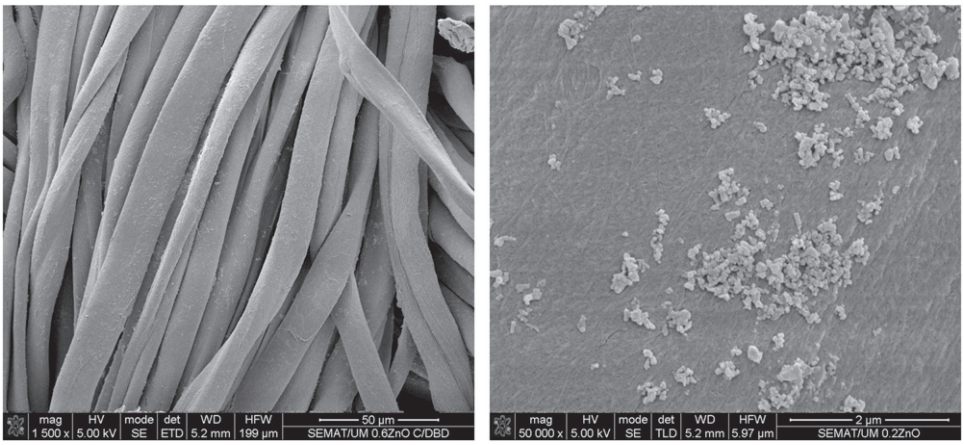

Figure 3. SEM images (X1500 and X50000 magnification) of antimicrobial ZnO NPs—PNIPAM composite coated on DBD plasma pre-treated cotton fibers. Images were carried in a FEG-SEM, NOVA 200 Nano SEM, FEI Company. Secondary and backscattering electron images were performed with an acceleration voltage of 5 kV and 15 kV, respectively. Samples were covered with a film of Au–Pd (80–20 wt.%) in a high-resolution sputter coater, 208 h Cressington Company, coupled to a MTM-20 Cressington High Resolution Thickness Controller.

mechanisms. In addition, penetration of the cell envelope and disorganization of bacterial membrane upon contact with ZnO NPs were also indicated to inhibit bacterial growth. However, the role of the Zn^{2+} ion released from the dissolution of ZnO is not yet clear, and the antibacterial mechanism of ZnO is still under investigation [90].

Nanocopper

Limited information, almost exclusively on cotton, is available on the antimicrobial activity and action mechanism of nano CuO in textiles [38, 55, 56, 91–93]. CuO is cheaper than silver, easily mixed with polymers and relatively stable in terms of both chemical and physical properties. Very recently, Teli *et al* have developed a bamboo rayon fabric grafted with acrylamide utilized to immobilize copper NPs. The product showed antibacterial activity against Gram-positive and Gram-negative bacteria and was found to be durable until 50 washes [94]. Perelshtein *et al* has recently sonochemically coated a cotton fabric with CuO NPs while maintaining antibacterial properties even after 65 cycles of washings according to hospital protocols of hygienic washing (75 °C) [95]. However, in comparison with AgNPs, higher concentrations of CuO are required to achieve a comparable bactericidal effect [96]. Moreover, CuO NPs synthesis is often more challenging in comparison to noble metals such as silver and gold. Copper sulphate in aqueous solution tends to form Cu_2O due to the relatively low CuO/Cu^{2+} redox potential and spontaneous oxidation of the NPs in ambient conditions. This last drawback can be avoided by protecting copper NPs against oxidation during preparation and storage using non-aqueous solvents, surfactants or ligands to prevent NP agglomeration during the process of synthesis [97].

Other metals and metal oxides

Very few examples are found in the existing literature about the use of other nanomaterials in textiles for antibacterial purposes. Gouda *et al* has used *in situ* synthesized zirconium oxide NPs deposited into cotton gauze fabrics. ZrO_2 NPs gave a 98% and 95% reduction rate in colony count against Gram-positive and Gram-negative bacteria, respectively. However, antifungal activity was lower than that of fabrics treated with nanosilver. No skin irritation was observed, and all prepared samples were durable enough to wash even after 30 laundering washing cycles [98]. Tang *et al* developed a simple *in situ* synthesis route for gold NPs to be applied to multi-functionalized silk fabrics. The AuNPs were prepared in a heated solution containing white silk fabric samples. Silk fabrics treated with AuNPs showed strong antibacterial activity, excellent UV protection properties and enhanced thermal conductivity. However, silk fabrics were colored red and brown by the AuNPs because of their localized surface plasmon resonance property [99]. Harifi *et al* prepared multi-functional polyester fabric with magnetic, antibacterial and sono-Fenton catalytic activities by *in situ* synthesis of magnetite and hematite NPs using ferric chloride, ferrous sulphate and sodium hydroxide. The results suggest the potential of the proposed method in producing fabrics with durable magnetic properties that are suitable for various applications such as electromagnetic shielding, antibacterial fabrics and sono-Fenton catalyst for dye discoloration [100]. In their review, Dastjerdi and Montazer discussed other nano-structured, antimicrobial agents with a potential for textile modification, including carbon nanotubes, nanoclay and its modified forms, and gallium- and liposome-loaded NPs; however, no textile applications have yet been developed [32].

Nano-additivated fibers and nanofibers

Several methods also include the bulk modification of conventional filament yarns of polyamide or polypropylene with various concentrations of different nanocomposite fillers, such as Ag, chitosan, PVA, ZnO, TiO_2 and mixed Ag/TiO_2, via melt mixing [86, 101, 102]. Yeo and Jeong, for example, produced bi-component, sheath-core fibers prepared by using a melt–spinning method with polypropylene chips and AgNPs. However, the fibers containing AgNPs in the core part showed no antibacterial activity. Only fibers having AgNPs in the sheath part showed antibacterial activity [103]. On the other hand, Dastjerdi *et al* produced biostatic polypropylene filament yarns with various blending contents of nanocomposite based on Ag/TiO_2 NPs using a twin-screw extruder. However, despite having good biostatic properties, none of the tested blends displayed a bactericide effect [78]. The bulk modification of filament yarns with various concentrations of nanocomposite fillers via melt mixing is an environmentally friendly and easily adjustable modification method. However, it is limited to synthetic fibers, and the particles situated in the central part of the filaments hardly contribute to the fibers' antibacterial properties. Although the production of core–shell bi-component fibers can be helpful in removing this disadvantage, the required systems are not easily adaptable to industrial standards. A similar problem is also noticeable in the case of reduction of metallic salts to NPs in the bulk polymeric matrix [32].

These problems, however, could be solved by the use of electrospun nanofibers due to their high surface-area-to-volume and length-to-diameter ratios (figure 4) [73, 79, 104–112]. Electrospinning is a process carried out at room temperature that allows the production of

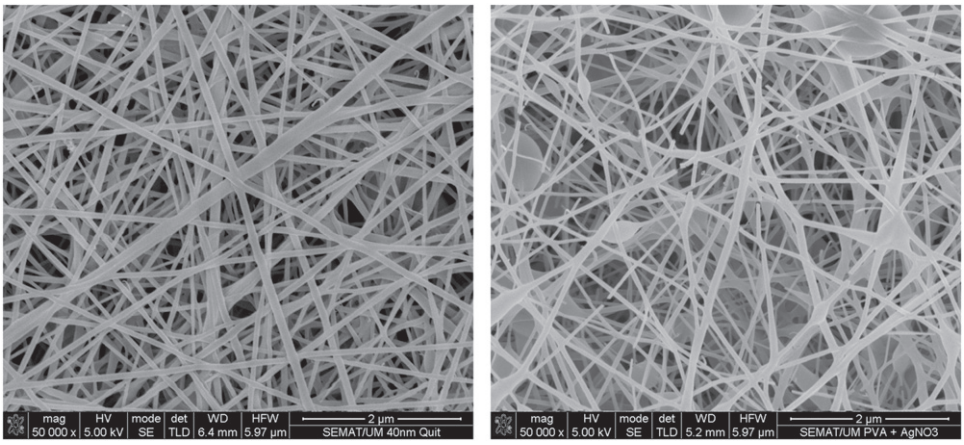

Figure 4. SEM images (X50000 magnification) of antimicrobial nanofibers obtained from PVA and chitosan (left), and PVA and $AgNO_3$ (right). Images were carried in a FEG-SEM, NOVA 200 Nano SEM, FEI Company. Secondary and backscattering electron images were performed with an acceleration voltage of 5 kV and 15 kV, respectively. Samples were covered with a film of Au–Pd (80–20 wt.%) in a high-resolution sputter coater, 208 h Cressington Company, coupled to a MTM-20 Cressington High Resolution Thickness Controller.

polymer fibers with diameters in the sub-micron size range, through the application of an external electric field, keeping intact the bulk properties of the polymers. Because of unique properties such as a high surface-to-volume ratio, very good mechanical performance, high porosity and diameters in the nanoscale, electrospun mats made from ultrafine polymer fibers have been drawing great attention for antimicrobial coatings. Moreover, electrospinning is a high quality, environmentally friendly and easily adjustable method for industrial applications. Several researchers have investigated the spinnability of different polymers. For instance, electrospun nanofibers of cellulose acetate, PVA, PAN and polyester urethane were used to disperse several antimicrobial materials, such as spherical gold and AgNPs, $Fe2O_3$, gallium nitride, zirconium carbide and carbon nanotubes [113–115]. Suspension of AgNPs directly combined into electrospinning polymer solutions is the most used method to prepare composite nanofibers. However, nanofibers produced using this method have demonstrated diminished antimicrobial efficiency due to nanoparticle aggregation. A more efficient method was the *in situ* reduction of silver ions in pre-electrospinning solutions, resulting in a more uniform dispersion of AgNPs [116].

Electrospun nanofibers based on chitosan and chitosan NPs applied on several textiles such as cotton, viscose and polyester fabrics have also been extensively investigated. Solutions of pure chitosan are not electrospinnable, independently of their polysaccharide concentrations, mainly due to the high surface tension and conductivities of chitosan acetic acid solutions. Electrospun antimicrobial nanofibers may, however, be fabricated from blended systems of chitosan and fiber-forming polymers such as nylon, cellulose acetate, PEO, PET, PAN and PVA [117–119]. Electrospinning allows extensive tunability in material properties and functions through the selection of polymeric nanofibers, ceramic nanofibers, metallic nanofibers or composite nanofibers. Ideally, nanofibers should be made into continuous yarn before weaving into textile fabrics. However, the diameter of the yarn collected using this process was less than 5 μm and it is uncertain whether the yarn was strong enough to be woven into textiles since

several studies showed that insufficient nanofibers in the bundle would result in yarn breakage [120]. For these reasons, now the majority of the electrospun nanofibers incorporating antimicrobial properties are utilized for the production of filtration membranes in order to reduce the formation of biofilm, which is a common source of membrane fouling.

Chitosan

Due to the environmental and toxicological concerns about the use of heavy metals for the production of NPs, researchers have been recently exploring the use of natural polymers, especially chitosan, in the antimicrobial finishing of textiles. Chitosan [$(C_6H_{11}O_4N)n$], the N-deacetylated derivative of chitin [$(C_8H_{13}O_5N)n$] due to the presence of amino groups, is a cationic polyelectrolyte, one of the few occurring in nature. This gives chitosan singular chemical and biological characteristics, such as biocompatibility, antibacterial properties, heavy metal ion chelation ability, gel-forming properties and hydrophilicity. The use of chitosan NPs in protein and drug delivery systems is being actively researched and reported in the literature [121]. However, the research on chitosan NPs for textile applications is limited because most of the literature is based on the use of bulk chitosan as a coating or finishing agent. Antimicrobial fabrics with nanocoated chitosan have proved to be a durable, cost-effective and eco-friendly process. Some research has shown, however, that chitosan NPs have a less inhibiting effect on *S. aureus* compared to bulk chitosan since NPs have less positive charge available to bind to the negative bacterial cell wall. Conversely, other researchers reported that chitosan NPs exhibit higher antibacterial activity due to the NP's larger surface area and higher affinity with bacteria cells, which yield a quantum-size effect [53, 122–125]. These contradictory results suggest that the antimicrobial mode of action of chitosan is not a simple mechanism, but is an intricate event-driven process that needs further investigation [126].

Conclusions

Most of the literature about antimicrobial textile nanocomposites is focused on silver. However, other metals and metal oxides such as zinc, titanium, copper, zirconium, iron and gold show improved biocidal properties at nanoscale. ZnO and CuO nanocomposites display similar performance compared to silver while TiO_2 efficacy is limited by light availability due to its photocatalytic mechanism of action. Despite the heterogeneous range of methods, textile substrates, nanoparticle sizes and concentrations that can be found in the literature, some general assumptions can be made about metal and metal oxide NPs based on the collected data. Silver and copper NPs of between 1 and 15 nm showed the best biocide activity at relatively low concentrations on the fabrics (5–50 ppm or 1–2 wt.%). AgNPs of up to 50 nm, still require relatively low concentrations of around 100 ppm or 5 wt.% to have complete Gram-positive and Gram-negative inhibition effects. Titanium oxide NPs applied to textiles are generally in the size range of 1–20 nm. With some exceptions, TiO_2 NPs showed low antimicrobial activity (an average of 70%) even at high concentrations of 10 wt.% on the fabrics. This occurs mainly because TiO_2 NPs are fully effective just under UV rays, which limits their practical use in the textile industry. On the other hand, ZnO NPs need a higher average size of 30–40 nm, but with a lower concentration (around 1 wt.%) than TiO_2 to be effective. This is possibly due to the synergetic dual effect of the photocatalytic generation of hydrogen peroxide and the direct

disorganization of the bacterial membrane. However, despite the promising results, the available information of ZnO NPs on textiles is still limited. All types of metal and metal oxide NPs with diameters greater than 100 nm need concentrations comparable to the metal ions or bulk materials to achieve the same antimicrobial performance.

The bacterial species *Staphylococcus aureus* (Gram positive), *Escherichia coli* (Gram negative) and *Klebsiella pneumoniae* (Gram negative) are the most-tested strains. Some authors have also tested different bacteria and fungi such as *Candida albicans*. However, the eukaryotic cytotoxicity and allergic reactions in humans are not considered in NP-containing textiles. Moreover, few authors have tested the antimicrobial efficiency after a reasonable number (at least 20) of washing cycles, limiting the precise estimation of the amount and form of NPs released from the fabrics into the environment. The risk assessment of the nanomaterials used in commercial textile products requires a better understanding of nanomaterial mobility, bioavailability and toxicity in the environment. Due to this increasing dichotomy between environmental and health concerns and the potential benefits of using NPs as an antimicrobial finish for textiles, the use of natural polymers, especially chitosan, and electrospun nanofibers have been recently explored. The research about chitosan NPs deposited on fabrics is still at an early stage; however, from the little information available, it is possible to estimate that the average sizes range from 20–200 nm and that the effective concentration is usually lower than 1 wt.%. The latest research in this field seems to indicate an emerging new paradigm in the production and distribution of NPs for textile applications utilizing non-toxic renewable biopolymers such as chitosan, alginate and starch.

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Author contributions

Andrea Zille performed the AgNP synthesis, AgNP deposition on polyamide fabrics, SEM and TEM analysis of the AgNPs and nanofibers, analyzed the data, interpreted results, elaborated table 1 and wrote the review. Noémia Carneiro performed the ZnO NPs synthesis, ZnO NPs deposition on cotton fabrics, SEM analysis of the deposited ZnO NPs, analyzed data, interpreted results, help in the table elaboration and reviewed the manuscript. António Pedro Souto performed the DBD plasma pre-treatment of the fabrics, analyzed data, interpreted results, helped with the table elaboration and reviewed the manuscript. Carla J Silva has prepared the electrospun nanofibers. Luís Almeida, Teresa Amorim and Maria Fátima Esteves analyzed data, interpreted results, helped with the table elaboration and reviewed The manuscript.

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