Synthetic Antimicrobial Agents for Textile Finishing

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Textiles with antimicrobial functionality have been intensively and extensively investigated in the recent decades, mostly because they are present in everyday life in various applications: medicine and healthcare, sportswear, clothing and footwear, furniture and upholstery, air and water purification systems, food packaging etc. Their ability to kill or limit the growth of the microbial population in a certain context defines their activity against bacteria, fungi, and viruses, and even against the initial formation of the biofilm prior to microorganisms' proliferation. Various classes of antimicrobials have been employed for these highly specialized textiles, namely, organic synthetic reagents and polymers, metals and metal oxides (micro- and nanoparticles), and natural and naturally derived compounds, and their activity and range of applications are critically assessed.

antimicrobial textiles | synthetic antimicrobial reagents | polymers

1. Introduction

1.1. General Background

Health risks management has been constantly considered in recent decades in all relevant domains in daily life due to the spectacular worldwide increase in number and variety of microbial infestation and proliferation, ranging from local to global, and from aggressive to violent and nonresponsive epidemics/pandemics (plague, SARS, West Nile, SARS-CoV-2, COVID-19, cholera, smallpox, scarlet rash, HIV-AIDS, Marburg, Ebola, Spanish flu, MERS) [1][2][3]. Thus, the employ of textiles with antimicrobial functionality has expanded up to unexpected rates. This market was estimated at USD 10.7 billion in 2021 and was projected to reach a 50% increase by 2026 at a compound annual growth rate (CAGR) of 6.5% in the same interval [4].

Subsequently, the scientific literature recorded an increase in the number of articles reporting on antimicrobial textiles and their specific finishing, reagents, and processing. A bibliometric analysis indicated in 2021 a number of publications of 534 articles per year [5], but the domain is very active and the rapid progress is abundantly documented by the most recent literature reports, which also illustrate the variety of new features connected to the subject [6][7][8][9][10][11][12][13][14][15][16][17][18][19][20][21][22][23][24][25].

Furthermore, recent surveys confirmed this trend. For example, data from the Web of Science Core Collection confirmed a number of 50 review articles published in the interval 2018–2023 on topics considered. Moreover, a significant number of patents—245—has been reported in the interval 2018–2023 (https://patents.google.com; accessed on 26 April 2023). Some of these data are illustrated in **Figure 1** and **Figure 2**, where the selection criteria are given in the legend.

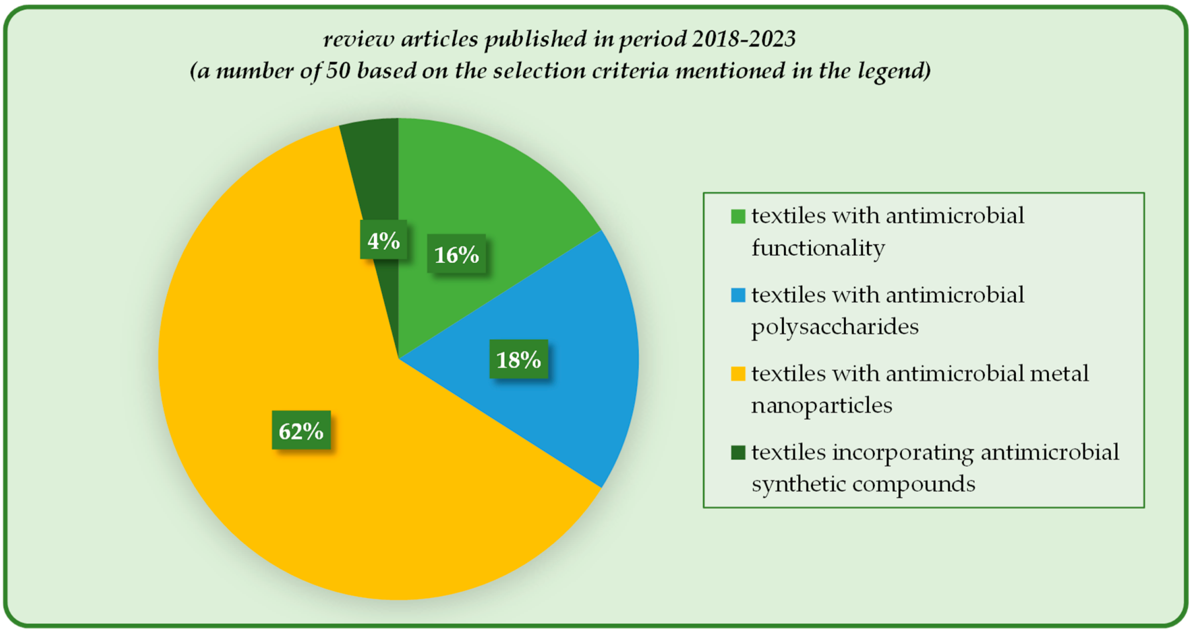


Figure 1. Review articles published in 2018–2023 (data from Web of Science Core Collection).

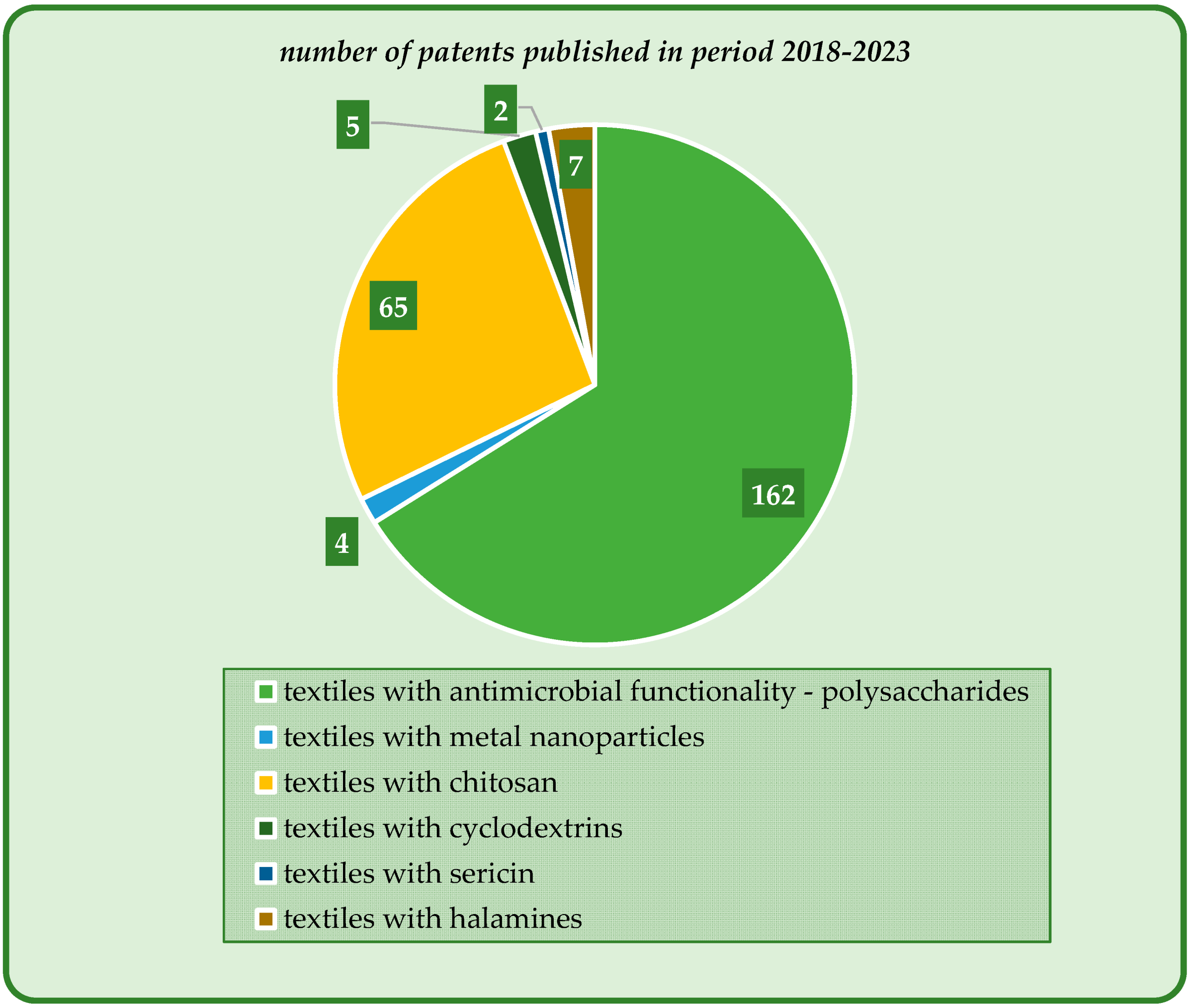


Figure 2. Patents published in the interval 2018–2023 (data acquired from https://patents.google.com; accessed on 26 April 2023).

First and foremost, textiles with antimicrobial finishing have to comply with several requirements: prevent, control, and/or eliminate microbial infestation, growth, and cross-infection over a wide spectrum; reduce odor, prevent staining, and maintain freshness for long intervals; must be stable, safe, durable, and reusable (in certain applications) [26]. Considering their antimicrobial effectiveness and the mechanism of action, as well as their toxicity versus tolerance, nature of fibers, and durability, textiles with antimicrobial functionality may be divided into several classes [18]:

- biostats, biocides (antibacterial, antifungal, antiviral), barriers, and antibiofilm;
- textiles with bound or leaching antimicrobial finishing;
- textiles made of natural (cotton, wool, silk, linen) or synthetic fibers (PP, PE, PES) or blends (cotton/elastane, cotton/PES, wool/acrylic);
- textiles able to release compounds with biologic activity;

- wearable and washing resistant.

Commonly, microorganisms are divided into different classes: bacteria, archaea, protozoa, algae, fungi, viruses, and multicellular animal parasites [27]. They have distinct features; most of them do not negatively interfere with human biota, but some can be or become pathogenic when certain favorable conditions are met. Bacteria are mainly divided into Gram-positive (*Staphylococcus aureus*) and Gram-negative (*Escherichia coli*). Other pathogenic bacteria (*Plasmodium malariae*, *Mycobacterium tuberculosis*, *Clostridium tetani*, *Corynebacterium diphtheriae*, *Treponema pallidum*), fungi (*Cryptococcus neoformans*, *Candida auris*, *Aspergillus fumigatus*, *Candida albicans*, *Candida glabrata*), and viruses (Ebola, herpes, hantavirus, papillomavirus, HIV, COVID-19) of particular concern have been used to evaluate the level of performance of antimicrobial textiles.

1.2. Processing Techniques

Textiles with antimicrobial functionality are materials of high interest; therefore, their processing is a key factor in their activity and stability. Padding, spraying, grafting, and cross-linking are some of the most relevant techniques. However, the development of biocide/biostatic textiles made of synthetic fibers has allowed new methods, such as compounding extrusion and melt blending [28][29]. At the same time, the employ of colloidal solutions, plasma treatments, magnetron sputtering, sol–gel processes, microencapsulation techniques, or even *in situ* formation/growth of different antimicrobials onto textile supports are modern processing methods that grant textiles enhanced activity and stability [28].

Coating is one of the most popular procedures and is suitable for both yarns and fabrics, natural and synthetic fibers, knitted, woven, and nonwoven textiles. Direct coating can be achieved by knife, roller, or calendaring, and the finishing must be viscous in order to form a satisfactory coating. The spray-coating technique uses an airbrush and the finishing solution must be less viscous. The method may be applied to nanoparticles deposition as well [30].

The exhaust method was “imported” from dyeing processing and comprises the transfer of the active reagent from a batch to the textile substrate, sometimes in the presence of a binder, and a curing stage is required to stabilize the coating. Thiazole-derived reagents have been successfully applied by this method to textiles which subsequently exhibited high effectiveness against Gram-positive and Gram-negative bacteria [31].

The pad–dry–cure approach, also known as the mechanical thermal fixation or padding, is suitable for micro- and nanoparticulate coating materials with low or no affinity toward the textile substrate. The thermal treatment must be short (1–5 min) and at high temperatures (100–150 °C) in order to reach an appropriate cross-linking degree (thermal fixation). The method is simple and effective [28].

Textile substrates may be submitted to different methods of surface modification in order to achieve better compatibility with the antimicrobial finishing reagents. Plasma techniques, microencapsulation and ultrasound methods are among the most employed.

Plasma treatments are highly effective and environmentally friendly, despite their drawbacks (high-energy-consuming process, expensive equipment), and are used to clean/etch or create new functional groups onto textile surfaces, to deposit thin films of nanometric thickness, or even grow nanoparticles *in situ*. The possibility to limit the in-depth alteration of the support is considered the main advantage of this method because it prevents the alteration of the bulk properties of the textile [21][32]. Plasma grafting and polymerization can be applied to a wide range of antimicrobial finishing reagents (quaternary ammonium salts derivatives, dichlorophenol, triclosan, chitosan, guanidine-based compounds, metal and metal oxides nanoparticles) when natural, synthetic, or blended textiles are used as support [21]. Plasma and magnetron sputtering were preferred for metals and metal oxides nanoparticles deposition (Ag, Ti, Cu) onto different substrates when stable coatings were obtained [9][33][34]. Moreover, it was recently reported that the emergence of highly effective antiviral textiles for personal protective equipment was favored by the employ of plasma processing [35][36].

The microencapsulation technique is a modern method used to manufacture antimicrobial textiles, having the advantage that the core is protected and thus the degradation under the action of external factors is prevented. Moreover, the microcapsules are stable and safe to handle and apply to the textile support [37][38]. The approach is preferred when natural and naturally derived compounds are used as antimicrobial finishing reagents. It can be achieved by chemical (*in situ* polymerization in oil-in-water emulsion; interfacial polymerization) and physico–chemical (coacervation, molecular inclusion complexes) methods, and the obtained coatings are resistant to friction, sunlight, washing, and wet/dry cleaning [39].

Nanotechnology is also employed in the manufacture of antimicrobial textiles in various manners. The sol–gel method is a wet chemical procedure and uses colloidal solutions of monomers as precursor to form an interpenetrated network with the textile support or to deposit particles onto the textile surface [28][40]. Metals and metal oxides can also be applied onto textiles by this method, as in the case of titanium dioxide and zinc oxide nanoparticles used for coating fabrics able to prevent the spreading of nosocomial infections [41] or for textiles with antibacterial activity and self-cleaning properties [42]. Cotton, wool, and silk fabrics are suitable for this method and a wise selection of reagents for the sol phase can impart in the end a multiple functionality to the textiles, alongside their biocide activity [28].

In situ synthesis of nanoparticles has the advantage of nanoparticles deposited directly onto the textile support, rather homogeneously, without binders or stabilizers, thus significantly reducing the waste and pollution (and the safety and environmental risks, respectively) and increasing the stability of deposition. Metals and metal oxides (Ag, ZnO, Fe, Au) are mostly used for this technique applied to natural or synthetic textiles [1][28][43].

Highly specialized fibers with antimicrobial activity have been successfully obtained by electrospinning, a modern technique that allows materials made of biopolymers or synthetic polymers, with fibrous/porous morphology, and having tailored biocide properties [44][45].

In the following, some new trends and advances in the field of highly specialized textiles with antimicrobial functionality are presented, as illustrated by recent reports.

2. Synthetic Antimicrobial Agents for Textile Finishing

Antimicrobials encompass a large variety of chemical compounds and physical agents that act on microbes (bacteria, fungi, viruses, protozoa) in general. They are used to kill bacteria or to prevent their development. However, many of them exhibit some serious drawbacks that restrict or prohibit their use, such as the emergence of resistance developed shortly after their introduction, and undesired side effects. At the same time, chemical biocides are potentially harmful substances for the environment and human health if not handled or processed properly.

N-halamine compounds are organic biocides capable of killing microorganisms without releasing free oxidative halogen until they come into contact with microorganisms. They present efficiency against a broad spectrum of microorganisms, long-term stability, non–toxicity to humans, regenerability upon exposure to aqueous free chlorine solutions, and excellent biocompatibility. In addition, microorganisms do not develop resistance to this class of antimicrobials. The surface of the materials influences the antibacterial mechanism of *N*-halamines and has an important role in their antibacterial effectiveness. A large number of places of contact with bacteria increases the inactivation rate and is favored by a larger surface area.

N-halamine biocides have been used in different applications such as water filtration systems, disinfectants in pools, textiles, and medical devices [46]. *N*-halamines and some other synthetic compounds, such as quaternary ammonium compounds, polyhexamethylene biguanide, and triclosan, have been applied for antimicrobial treatment of textiles. Antimicrobial fabrics have found different applications in pharmaceutical, medical, engineering, agricultural, and food industries [47][48]. The *N*-halamine-treated fabrics can be rendered as having excellent antimicrobial activity through a bleaching process and can inactivate a broad spectrum of microorganisms, including Gram-negative and Gram-positive bacteria, in relatively short contact times. When the oxidative halogen is consumed, textiles modified with *N*-halamines regain their antimicrobial properties by exposing them to diluted household bleach. However, the practical application of *N*-halamines involves some disadvantages. For example, the cost of the treatment increases in the case of the use of organic solvents necessary to dissolve some *N*-halamine derivatives, which also presents safety risks.

As surfactants, quaternary ammonium compounds concentrate at the interface between the lipid-containing bacterial cell membrane and the surrounding aqueous environment. There are two types of interaction between quaternary ammonium salts and microbes: a polar interaction, occurring by cationic nitrogen, and a non–polar one, attributed to the hydrophobic chain. The cationic ammonium group can interact with the negatively charged cell membrane of bacteria. This attraction force induces the generation of a surfactant–microbe complex which can interrupt the activity of proteins, including all of the important functions in the cell membrane and even bacterial DNA. Furthermore, hydrophobic groups can penetrate into the microorganism and cancel all of the key cell functions. Increasing the length of the alkyl chain results in increasing the antibacterial activity of quaternary ammonium salts [49].

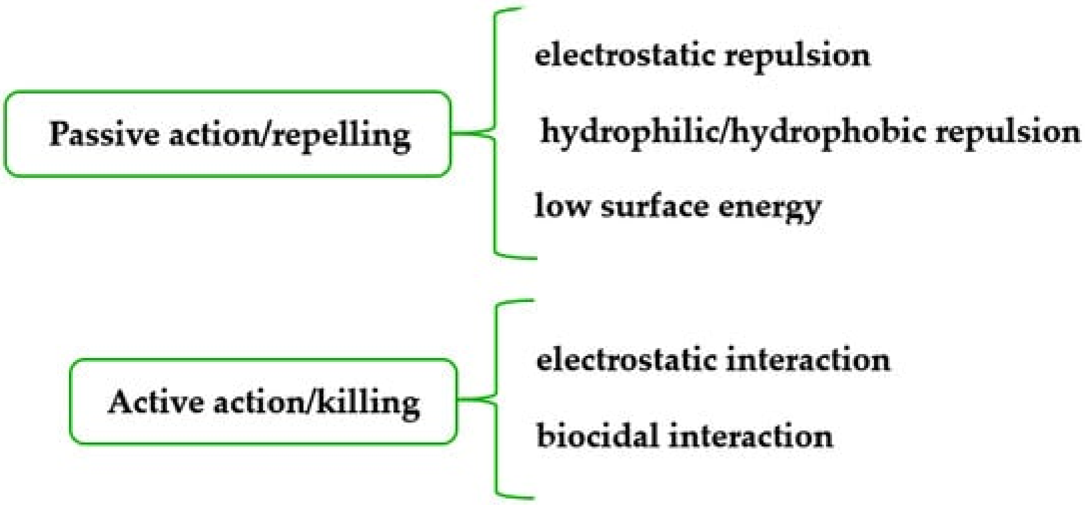
Quaternary ammonium compounds have no effectiveness against difficult-to-kill non–enveloped viruses. Among the extremely effective disinfectants with a wide spectrum and short contact times (3–5 min), they can count the formulations with low alcohol content used against bacteria, enveloped viruses, pathogenic fungi, and mycobacteria. Disinfectants based on quaternary ammonium salts with the addition of alcohol or solvents bring about a much faster drying of the products on the applied surface, which results in an ineffective or incomplete disinfection. In addition, quaternary ammonium compounds kill algae and are used in industrial water systems to counteract unwanted biological growth. Cetrimide (alkyltrimethylammonium bromide) and benzalkonium chloride have antibacterial, antifungal, and antiviral (enveloped viruses) properties and can be applied to the skin or mucous membranes to avoid or minimize the risk of infection. Hard water, anionic detergents, and organic matter reduce the activity of these disinfectants based on quaternary ammonium salts, which is a disadvantage. Moreover, *Pseudomonas* can metabolize cetrimide, using it as a carbon, nitrogen, and energy source.

Triclosan has antiseptic and disinfectant properties and a significant action against Gram-negative and Gram-positive bacteria. The acaricide benzyl benzoate in its structure accounts for protection against mites and it is used in acaricide (spray or powder) formulas, and for the treatment of scabies as a solution (25% concentration). Triclosan has been widely used in a large number of consumer products, such as cosmetics, toothpastes, deodorants, soaps, toys, and surgical cleaning treatments, based on its non-toxicity and antibacterial properties. Although triclosan is not considered to be as toxic as other pollutants, its occurrence in wastewaters, biosolids, and aquatic and terrestrial environments remains a concern. Furthermore, triclosan exhibits certain physicochemical characteristics that make it difficult to remove from the environment. There are studies that attribute some harmful health effects to triclosan, such as skin irritation, hormonal disruption, interference with muscle function, and contribution to antibacterial resistance [50].

Chlorhexidine has a cationic molecular component that attaches to negatively charged cell membrane area and causes cell lysis. As an antiseptic, chlorhexidine is used as a mouth rinse and endodontic irrigant due to long-lasting antimicrobial effect attributed to its binding to hydroxyapatite. It is commonly held that chlorhexidine would be less caustic than sodium hypochlorite. Similar to sodium hypochlorite, heating chlorhexidine in low concentration increases its local efficacy in the root canal system and maintains low systemic toxicity. Chlorhexidine presents drawbacks, such as its incapacity to dissolve necrotic tissue remnants and chemically clean the canal system, and lower effectiveness on Gram-negative than on Gram-positive bacteria [51].

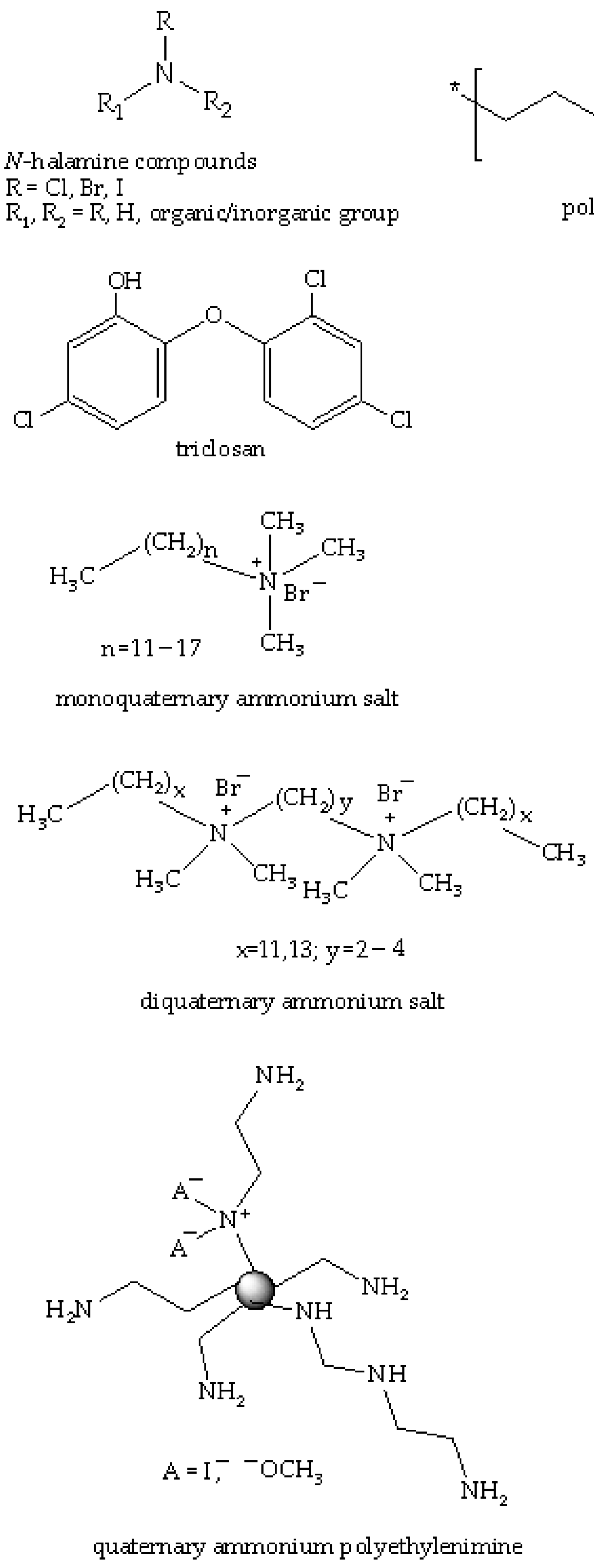
Common antimicrobial agents are prepared from natural or low-molecular-weight compounds. Due to biocidal diffusion, they present toxicity to the human body. In addition, they are easily susceptible to resistance and can lead to environmental contamination. Antimicrobial polymeric materials can overcome these problems by promoting antimicrobial efficiency and reducing residual toxicity. Moreover, antimicrobial polymers exhibit chemical stability, non–volatility, and long-term activity. Polymers containing covalently linked antimicrobial moieties avoid the penetration of low-molecular-weight biocides from the polymer matrices, unlike antimicrobial polymers obtained by physical methods (trapping or coating of organic and/or inorganic active agents during or after processing). These antimicrobial polymers are environmentally friendly and show durability over time. The most studied antimicrobial polymeric materials, and probably the most used, are those based on quaternary ammonium and/or phosphonium salts [52]. In addition, polymeric *N*-halamines with or without reactive functional groups were used to coat different fabrics by various approaches [49].

During the last two decades, synthetic (co)polymers have been designed to mimic the prominent physio–chemical characteristics of host defense peptides. Although these polymers have revealed a broad-spectrum antimicrobial activity, rapid bactericidal kinetics, and a very low propensity to induce resistance, none of them has been currently in clinical trials [53]. The schematic reaction mechanism of passive and active action of the antimicrobial polymers is presented in Scheme 1.



Scheme 1. The mechanism of action of antimicrobial polymers.

Concerning the conducting polymers, namely, polyaniline, polypyrrole, and polythiophene, their biomedical applications have not been well studied even though they have good antimicrobial activity. This limitation may be dampered by the preparation of polymer blends and nanocomposites with different (bio)polymers and nanomaterials, respectively, to achieve the desirable biocompatibility and physicochemical properties [54]. **Table 1** summarizes the most relevant antimicrobial agents presented above, their applications, and mechanism of action, and Scheme 2 illustrates the chemical structures of the most important antibacterial compounds.



Scheme 2. Chemical structures of some conventionally used synthetic antimicrobial agents.

Table 1. Synthetic antimicrobial products, their applications, and mode of action.

Antimicrobial Agent	Properties and Applications	Antimicrobial Mechanism	Ref.
Quaternary ammonium compounds Polymeric materials having onium salts (quaternary ammonium and/or phosphonium salts) Quaternary ammonium polyethylenimine	- Healthcare, household products, surface preservation, food industry, pharmaceutical/cosmetic (preservation) - Highly effective as antimicrobial agents in orthodontic cements to introduce antibacterial activity toward *S. mutants* and *L. casei*	The long, lipophilic alkyl chain of the quaternary ammonium compounds perforates cell membranes, and produces the release of cytoplasmic components, autolysis and cell death of the microbial strain	[52] [55] [56] [57] [58] [59]
Halogenated phenols Triclosan	- Antiseptic, disinfectant, fungicide, pesticide, antimicrobial, antiseptic, preservative - Antimicrobial activity against many types of Gram-positive and Gram-negative non- spore-forming bacteria, some fungi - Clinical settings, consumer products (cosmetics, cleaning products, paint, plastic materials, toys) - Durable antifungal finishing of cotton fabrics	Inhibits the active site of enoyl-acyl carrier protein reductase enzyme, which is essential to the fatty acids synthesis of bacteria and the building of the cell membrane	[10] [58] [60] [61]
Chlorhexidine Hexametaphosphate salt of chlorhexidine (as nanoparticles) Polyhexamethylene biguanide (PHMB)	- Preoperative skin cleansing preparations, hand disinfectants, and oral mouth rinses - Efficient antimicrobial agent against gram-negative and -positive bacteria and yeasts. - Biomedical materials and consumer products - Antimicrobial efficacy against MRSA and *P. aeruginosa*, in both planktonic and biofilm growth conditions - Healthcare uniforms - Nonspecific antimicrobial properties and remained efficient (>99% against *S. aureus* and *K. pneumoniae*) after use for 5 months	Chlorhexidine inhibits membrane-bound ATPase, based on cell membrane disruption and leakage of intracellular constituents, a rapid process with most damage occurring within 20 s of exposure The positively charged biguanidines bind to negatively charged phosphate group of the bacterial cell wall or virus envelope, breaking the membrane integrity, which leads to cell lysis and subsequent cell death	[25] [62] [63] [64]
N-halamines	- Antimicrobial activity against a broad spectrum of microorganisms, rechargeability, nontoxicity to humans - Medical devices, water purification, hospitals, antibacterial modification of cotton fabrics	The direct transfer of oxidative halogens to a cell after contact resulting in oxidation of the amino acids in the cell membrane and inactivation the microorganism	[46] [49] [65] [66] [67] [68]

Antimicrobial Agent	Properties and Applications	Antimicrobial Mechanism	Ref.
	- Antimicrobial activity against aerosolized bacteria - Air filtration technology - Biocidal properties against *S. aureus* and *E. coli* - Food packaging and biomedical applications		
5,5-dimethylhydantoin	Cotton fabric with regenerable antibacterial properties against *S. aureus*	Coating dimethylhydantoin on cotton fabric (by pad–dry–plasma–cure process) followed by chlorination inhibits the bacteria	[69]
Cinnamic acid derivatives	Pharmacological, antifungal, and antibacterial action	Plasma membrane disruption, nucleic acid and protein damage, and the induction of intracellular reactive oxygen species	[70] [71] [72]
Polyaniline and its derivatives	- Bacteria-resistant surfaces against *S. aureus* and *E. coli* - Wall and room-door coatings in hospitals	Different oxidation states of polyaniline and presence of functional groups	[73]
Polypyrrole (nanoparticles)	Antimicrobial treatment against *S. aureus* and *E. coli* of polyester fabrics	The positive charges (=NH+) in the polypyrrole backbone that are created by dopant compounds	[74] [75]
Polythiophenes	Antimicrobial compounds able to kill bacteria selectively by damaging negatively charged cell envelopes	Cationic charges with capacity to create huge amounts of singlet oxygen that interact with organism	[76]

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