

Recent Advances in Antibacterial Composite Coatings

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For the removal of pathogens, classical methods such as chemical disinfection, sterilization (thermal or ionic) are used and continue to be used, but the current conditions of energy conservation and environmental protection require approaching this problem from a different perspective.

Lately, the current trend is to design new materials that, by coating with different substances or by functionalizing surfaces, ensure the necessary antimicrobial protection by diminishing the adhesion of microorganisms to the surfaces or by reducing biofilm formation, persistence or proliferation of pathogens on various materials. This Special Issue brings to light the latest research contributions that could answer the two major questions: “What strategies can we choose to design materials with antimicrobial properties?” and “How does a material with antimicrobial properties act against different pathogen strains?”. Thus, this Special Issue gives the opportunity to have full access to a collection of original research studies that address the synthesis of emerging antibacterial intelligent materials that are tremendously necessary for a large spectrum of applications from medical, textiles, process separation and daily life products.

To give a brief answer to these two questions this Editorial will try to clarify some aspects in terms of classification of materials with antimicrobial properties and their interactions with pathogenic microorganisms. Afterwards, a description of different strategies for antimicrobial material design will follow-up, putting forth evidence for the efficiency of these materials for the targeted pathogen strain.

In the case of synthetic materials, which are sensitive to high temperatures for a proper cleaning, the defense against microbial development is missing and still represents a great challenge for product manufacturers that give a lot of attention to improving human health and reduce prolonged exposure to harmful microorganisms. Microbial cells connected to any artificial surface can thus survive and proliferate if the moisture conditions allow them. Over time, microbial cells that have adhered to the synthetic surface begin to build a biofilm consisting of a matrix of polysaccharides with integrated cells, which allows them to survive.

The current technological trend is directed toward the design of surfaces with intrinsic antimicrobial activity or to coating processes aimed at reducing of bacterial adhesion [1]. Antimicrobial surfaces are classified into antibiofouling and bactericidal surfaces. Antibiofouling surfaces do not allow the adhesion of bacterial cells on different surfaces by creating conditions that are not favorable for cell growth, while bactericidal surfaces are chemically modified surfaces that, due to certain functional groups of the chemical agents, interrupt the metabolism of bacterial cells that reach these surfaces. If it is not possible to create materials as such, with intrinsic antibacterial properties, an alternative is to cover the already existing materials with substances that create a continuous or discontinuous film with anti-adherence properties [1].

In the last decade, chemistry was a great leverage to design synthetic polymers with intrinsic antimicrobial properties, thus creating a new class of biocides that give an excellent alternative to current biocides and even antibiotics in some cases [2,3]. Their



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mechanism of action is sometimes not fully understood, but nevertheless, their great versatility allows them to possess a low capacity to develop resistant microbial strains. This property classifies them as eligible candidates for designing materials or covering surfaces with antimicrobial purposes.

In their recent work, Santos et al. [3] classified the antimicrobial activity of synthetic polymers based on their mechanism of biocidal activity in biocidal polymers, polymeric biocides, and biocide-releasing polymers. Thus, biocidal polymers possess intrinsic antimicrobial activity which is attributed to various cationic fragments such as quaternary ammonium, tertiary sulfonium, phosphonium and guanidium that exert antimicrobial effects by destabilizing the negatively charged cell membrane of microorganisms [3,4]. On the other hand, polymeric biocides contain repetitive bioactive units (i.e., amino, carboxyl or hydroxyl groups) covalently linked to the polymer chains that act through microbial repulsion or anti-adhesion rather than microbial killing actions [5]. Both biocidal polymers and polymeric biocides exert antimicrobial and antifouling effects, respectively, upon direct contact with a microorganism. Biocide-releasing polymer matrices loaded with the biocide compound act similarly with drug-release composites that liberate the biocide into the environment based on different control triggers (pH, temperature, etc.). This approach is sometimes preferred due to the possibility of releasing a large quantity of biocide at once, but is considered as one of the most harmful for the environment since these high concentrations are associated with toxic effects in the short term and less efficient in the long term [3–6].

Biocide releasing-polymers are nanoparticles most frequently synthesized from chitosan due to its special properties: antibacterial, pH controllable release, bio adhesion, biocompatibility and biodegradability. Other polymers used for biocide-releasing nanopolymer synthesis include polycaprolactone (PCL) and poly(lactic-co-glycolic acid) (PLGA) because they are inert and do not present major cytotoxic effects in mammalian cells [6]. Biocidal polymers, from the class of quaternary ammonium compounds (QACs), are present as classic and highly effective disinfectants and antiseptics in the fields of medical, pharmaceutical and personal care products, but some studies have indicated that their improper formulation could establish the development of resistant bacteria on surfaces treated with products that contain QACs at subinhibitory concentrations [7,8].

For this reason, to give an opinion about QACs or QACs products other strategies should be taken into account, such as: (i) new synthetic QACs development; (ii) revision the existing formulations for the QACs containing products; or (iii) consideration of other types of chemical compounds to ensure the desired antimicrobial or antifungal efficiency.

To enhance the antimicrobial activity of different surfaces or materials, a new generation of materials, namely zwitterionic polymers (formed with equal anion and cation groups on the molecular chains) [9], have been developed, due to their additional biocompatible and antifouling properties that are considerably important in the surgery field to reduce post-operative infections. Their synthesis strategy led to the development of polymers that mimic the activity of peptides on one hand and introduce antibacterial activity of the synthetic peptide on the other. Thus, the super hydrophilic characteristic of zwitterionic polymers allow the material to form a hydration shell through electrostatic interaction and thus confers the ability to prevent proteins adherence to medical implants, antifouling properties for membrane separation and applications in marine coatings [7,9,10].

Considering the new trends in “greener” approaches in designing new materials to address the regulations and directives that ensure environmental protection, the use of natural extracts that possess antimicrobial properties has gained significant attention in the last decade. Thus, the impregnation of different materials, even natural polymers (i.e., cotton fabrics, bacterial cellulose, poly-lactic acid, etc.), with natural antimicrobial extracts (i.e., thyme extracts, propolis, etc.) is a technique increasingly used nowadays, especially for packaging in the food industry or textile fibers to ensure protection against different pathogenic strains, such as *Escherichia coli*, *Staphylococcus aureus*, *Pseudomonas aeruginosa*, *Bacillus subtilis*, *Candida albicans*, etc. [11–16]. This technique is based on the physical

interactions between the active antimicrobial compound and the material that has the role of “supporting/entrapping” the active compound.

Furthermore, this technique has been applied to natural antimicrobial extracts and classical antibiotics in new emerging wound dressings with antimicrobial properties that could be used in the medical field [17–21].

Taking a step forward in probably what will be the future of food packaging, edible formulations have been taken into account in various research studies. Considering the multifunctionality of these modern materials, the antibacterial role of packages has to be considered in order to reduce food infestation and to extend the shelf life of food products. These ideas have started to become a continuous concern in the scientific world. Thus, there are some examples that include antimicrobial, biodegradable and edible packaging which involves the use of biopolymers, such as carboxymethyl cellulose, chitosan or polysaccharides modified with different natural extracts or inorganic nanoparticles with intrinsic antimicrobial properties showing efficiency against *Salmonella choleraesuis*, *Vibrio parahaemolyticus*, *Staphylococcus aureus*, *Diaporthe actinide*, *Penicilium expansum*, *Escherichia coli*, etc. [22–24].

Another method of functionalizing materials or surfaces in order to develop antimicrobial properties and prevent the formation of biofilms is doping them with different metals (Cu, Ag, Ce, Zn, Ti, Sr, etc.) in the form of inorganic nanoparticles with a broad spectrum of applications in the textile industry, construction field, membrane separation and medical applications [25–30]. This approach involves the impregnation method described above or the generation of inorganic nanoparticles directly on to the material’s surface previously functionalized with different chemical groups (i.e., carboxy, hydroxy, etc.) that favor the attachment of the inorganic nanoparticles [25–30].

In terms of designing antimicrobial surfaces, it is also worthy to mention coating strategies that have shown an efficient and eco-friendly manufacturing processes for the tailoring of functional materials or molecules attached to different substrates. This is especially desirable in the medical field for medical devices are sensitive to sterilization at high temperatures.

Singh et al., presented the advantages of a novel coating material derived from a catechol derivative dopamine, known as polydopamine (PDA), designed and developed with the ability to adhere to almost all kinds of substrates, mirroring the topographical features of some natural antibacterial materials [31]. Protein-based adhesives, secreted by marine mussels, contain the catechol amino acid, 3,4-dihydroxyphenylalanine (DOPA), which, in the presence of lysine, permits the ability to anchor them to various surfaces in both wet and saline habitats, being suitable for covering medical devices [31].

The natural compound tannic acid (TA), a tea stain polyphenol, has a good affinity for different substrates and is proven to be a great inhibitor of proliferation of pathogenic strains, making it a good candidate for the functionalization of different materials (i.e., polymers, nanomaterials, proteins, etc.) for antimicrobial surfaces or surgical sutures in the field of bioimplants [32,33].

Badica et al., demonstrated the antimicrobial activity of MgB_2 powders and their potential, when embedded in PVP-based polymeric coatings, for the fabrication of improved plastic medical devices, resistance to microbial colonization and thus, reduced probability to induce biofilm-associated infections [34].

Using innovative techniques, such as aerosol-assisted atmospheric pressure plasma deposition (AAPPD), Wang et al., studied the deposition of silver ions as nano-capsules on PET films, which are part of many medical devices, and their controlled release in order to obtain surfaces with antibacterial and antifouling properties [35].

Another field of antibacterial coatings refers to the coating of steel alloys, with the aim of obtaining civil structures (balustrades, surfaces, etc.) with antimicrobial properties, especially in areas of intense population traffic or resistant biocompatible structures found in public transport, malls, hospitals, educational units, etc. Thus, the latest strategies involve carbonaceous materials such as graphene oxide or carbon fibers dispersed into a polyetheretherketone (PEEK) matrix serving as a composite coating on Ti–6Al–4V alloys

for dental or orthopedic antimicrobial applications [36]. Another example involves the use of the antibacterial and anti-corrosive properties of nickel-graphene oxide (Ni-GO) and nickel-reduced graphene oxide (Ni-rGO) composite coatings against *S. aureus*. Here, the active material is deposited by magnetic field-assisted scanning jet electrodeposition on to manganese steel in order to obtain surfaces resistant to corrosion and with good antibacterial properties [37].

A similar goal has been obtained in the case of manufacturing a multifunctional composite coating on (Mg) AZ31 magnesium alloys through a micro-arc oxidation procedure using silver nanoparticles (AgNPs) as antimicrobial compounds and polyethyleneimine (PEI) as a medium for a self-assembled uniform coating of AgNPs for an antimicrobial barrier against *S. aureus* [38].

On the other hand, AISI type 316L stainless steel (SS) is a widely used material for processing equipment in the chemical, petrochemical and nuclear power industries, as well as for bio-implants due to its versatile mechanical, corrosion resistance and bio-compatible properties. To protect this alloy from biofilm formation a graphene oxide (GO)/polyvinylpyrrolidone (PVP) (GP) composite coating is deposited by electrophoresis on its surface and antibacterial tests prove its superior antibacterial performance against *Pseudomonas sp.* and *Bacillus sp.* and its antibiofouling properties [39].

Currently, the field of orthopedic and dental implantology requires resistant structures and coatings with antimicrobial properties necessary to modify/functionalize magnesium-based alloys used in biomedical applications. Thus, different structures impregnated with hydroxyapatite doped with different metals have been developed, such as copper-hydroxyapatite (HA) coatings [40], antibacterial composite materials based on the combination of polyhydroxyalkanoates (PHAs) with selenium and strontium co-substituted hydroxyapatite for bone regeneration [41], and the superhydrophobic composite coating of hydroxyapatite (HA)/stearic acid on magnesium alloys (AZ31B) [42].

On the other hand, a particularly interesting field for the study of antimicrobial coatings is that of naval and processing equipment that is immersed in water. Considering the conditions of prolonged contact with the aqueous marine environment, regardless of the temperature range, biofilm formation is inevitable. The presence of this biofilm can raise various technical and corrosive problems, which is why it is preferable that it is prevented from forming through different coating designs. This involves the change of the hydrophilic/hydrophobic balance of the final coating considering that in an aqueous environment in order to prevent biofilm formation hydrophobic or superhydrophobic coatings are necessary [43].

Some studies have assembled antibacterial hybrid multilayers onto NaOH etched basalt scales via mussel-inspired depositions of PDA and AgNPs followed by post-modification with 1-dodecanethiol. 1-dodecanethiol can suppress the formation of mature biofilms and maintain bacteria in a dispersed state, which further promotes the bacterial-killing effect due to the presence of AgNPs [43]. Additionally, Bi₂WO₆/boron-grafted polyurethane composite coatings (BWOB), composed of Bi₂WO₆ with three morphologies (nanosheet, flower and microsphere) and boron-grafted polyurethane, have been successfully synthesized to achieve highly efficient antifouling and antimicrobial properties against *E. coli*, *S. aureus* and *Nitzschia closterium* [44].

Another topical area with great antimicrobial coatings prospects is that of textiles used for protective equipment, sports equipment or for everyday clothing. Covering textile fibers with ingredients with an antimicrobial role is a challenge and complex situation to solve, considering that in addition to the role of antibacterial protection, they must protect the specific microflora of the human epidermis. Another drawback of creating feasible, antimicrobial clothes is related to the antibacterial efficiency after several washing cycles. Moreover, there is also the challenge of making clothes conductive, in order to communicate with electronic devices and store energy in clothes of the future while keeping their antimicrobial characteristics. Literature data has presented studies using different natural or synthetic fabrics with modified graphene oxide (GO) or reduced GO (rGO) to ensure both

electroconductive and antimicrobial properties [45]. In many cases, in order to amplify the intrinsic antimicrobial properties of graphene derivatives, different inorganic nanoparticles such as Ag, Fe, Cu, Zn, TiO₂, ZnO, CdS, MnS₂, etc. have been employed to design composite materials to prevent or kill *S. aureus*, *E. coli* or *P. aeruginosa* [45–47]. Enhanced effects have been obtained when using electroconductive polymers as coatings for different fabrics. The multifunctional material comprised of poly(aniline) deposited on polyester-viscose fabrics not only exhibits antimicrobial activity under acidic conditions against *S. aureus* and *S. epidermidis* bacteria, but also registers resistivities up to 10⁵ (Ω/cm) [48].

In conclusion, our Special Issue aimed to provide a forum for researchers to share current findings and to promote further research into the synthesis of advanced multifunctional and antimicrobial coatings, including new techniques, experimental models, new antimicrobial behaviors or theoretical calculations with a broad spectrum of applications.

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