Antibacterial Polymer Composite Materials

Subjects: Materials Science, Composites
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The escalating presence of pathogenic microbes has spurred a heightened interest in antimicrobial polymer composites tailored for hygiene applications. These innovative composites ingeniously incorporate potent antimicrobial agents such as metals, metal oxides, and carbon derivatives. This integration equips them with the unique ability to offer robust and persistent protection against a diverse array of pathogens. By effectively countering the challenges posed by microbial contamination, these pioneering composites hold the potential to create safer environments and contribute to the advancement of public health on a substantial scale.

Keywords: polymer composite ; antibacterial ; display coating ; sensor ; transparency

1. Introduction

The presence of harmful microorganisms in the surrounding environment can give rise to a wide range of social, economic, and human issues [1]. Indeed, the contamination of material surfaces by microbes plays a significant role in the rapid transmission of infectious diseases among individuals [2]. Interactive displays utilizing touchscreen technology have become prevalent in healthcare, public spaces, and industries, allowing easy information access through touch interactions in both public and semi-public areas [3][4]. However, these touchscreens serve as sources of various harmful pathogens, including bacteria, viruses, fungi, and parasites, which can readily spread to humans through the operation of such devices [5][6]. Specifically, the recent COVID-19 pandemic has heightened concerns regarding the use of touchscreen devices in public spaces, as they can be touched by multiple individuals, potentially facilitating the transmission of harmful pathogens and posing an infection risk [7][8]. A crucial strategy in disease prevention involves the regular cleaning and frequent disinfection of touchscreens. Nevertheless, conventional disinfection methods, which rely on chemicals like ethanol, isopropanol, and hypochlorite, are unsuitable for disinfecting touchscreens due to their sensitivity to these substances [9]. In addition to touchscreen devices, other surfaces, like door handles, elevator buttons, and escalator rails, in public areas can also become contaminated and pose a potential risk of the spread of microbial pathogens [10].

Commonly, polymers lack intrinsic antibacterial capabilities, necessitating innovative approaches to bestow them with antimicrobial properties. To this end, researchers have fervently explored avenues of modification and functionalization, seeking to augment polymers' ability to combat microbial threats. A particularly notable strategy involves the infusion of antimicrobial additives such as metals, metal oxides, and carbon-based materials into polymer matrices. This integration, while altering the composition, serves as a catalyst for bestowing antibacterial activity upon the resultant polymer composites. This methodology has garnered substantial interest across diverse scientific domains, owing to its potential to elevate the antimicrobial efficacy of polymers, thereby addressing crucial concerns associated with microbial contamination in various applications.

2. Metals-Incorporated Polymer Composites

Polymer composite materials infused with metals showcase antibacterial efficacy owing to the antimicrobial attributes of metallic nanoparticles. These nanoparticles can efficiently exterminate and impede the proliferation of bacteria on the surface of the composite material. The pivotal advantage of integrating these materials into composite coatings lies in their capability to deter the attachment of microbial pathogens while augmenting their antimicrobial potency [11]. Furthermore, polymers can enhance the mechanical attributes of composite films, which are crucial for display and sensor coating applications. Multiple studies have showcased the antibacterial effectiveness of polymer composites achieved through the integration of diverse metallic nanoparticles, encompassing silver, gold, copper, and zinc [12][13][14]. The antimicrobial efficacy of nanomaterials is intricately linked to their structural and physical characteristics, including size, shape, chemical composition, surface area, and zeta potential. The antimicrobial actions of metals encompass a range of processes, such as producing reactive oxygen species, releasing cations, inducing biomolecule harm, depleting ATP, and interacting with cell membranes, all of which contribute to the eradication of bacteria [15]. Silver is commonly chosen as one of the metals to be infused into a polymer matrix, thereby imparting the polymer with antimicrobial capabilities. Silver stands out as one of the most frequently employed metals for integration into polymer matrices to achieve antimicrobial effects [12]. For example, Hoque et al. demonstrated the dual function of polymer–silver nanocomposite with excellent antimicrobial activity against various bacteria and fungi. The preparation of the silver nanocomposites involved the biodegradable polymer N, N-dimethyl-N-hexadecyl ammonium chitin tosylate (Q-DMHC48) [16]. The surfaces coated with the nanocomposite demonstrated excellent antimicrobial activity in various drug-resistant bacteria, including methicillin-resistant *Staphylococcus aureus* (MRSA), vancomycin-resistant *Enterococcus faecium* (VRE) *P. aeruginosa*, and *Klebsiella pneumoniae*, as well as pathogenic fungi such as *Candida* spp. and *Cryptococcus* spp. In addition, nanocomposite-coated surfaces exhibited rapid killing and long-lasting antimicrobial activity, maintaining effectiveness over an extended duration. Moreover, when applied to catheters, the nanocomposites effectively reduced the burden on the catheter and in the tissues surrounding it in a mice model.

Several studies have also focused on the preparation of antibacterial polyurethane coatings impregnated with the incorporation of Ag nanoparticles, which are capable of releasing bactericidal silver ions upon contact with bacteria and fungi [17][18]. However, these coatings are generally effective only for short-term applications due to the high diffusivity of Ag NPs and their tendency to aggregate. Thus, Mohammadi et al. prepared a silver(I) complex with a Schiff base ligand (SBL)-extended waterborne polyurethane (WPUL/Ag) with high storage stability and a low aggregation tendency [19]. The WPUL/Ag composite coating exhibited strong antibacterial activity against both Gram-positive Staphylococcus aureus and Gram-negative *Pseudomonas aeruginosa* bacteria, achieving a bacterial reduction of 99.99%, while the WPUL revealed no antibacterial activity. Copper is inexpensive compared with other metals, and is a widely used material for preparing various antibacterial polymer composites [20]. For example, Pinto et al. prepared a cellulose-based biopolymer nanocomposite using copper nanostructures, namely nanoparticles and nanowires, and investigated its antibacterial efficiency against *S. aureus* and *K. pneumoniae* [21]. The composite with copper nanowires exhibited less antibacterial activity than the nanoparticle-based composite. Additionally, a significant improvement in antibacterial activity was observed with increasing copper content. In another study, antibacterial thermoplastic polyurethane (TPU) was prepared by incorporating 1 wt% copper particles through the melt blending method [22]. They observed the resulting composite films successfully hindered the growth of Staphylococcus aureus (*S. aureus*) and *Escherichia coli* (*E. coli*), effectively inhibiting biofilm formation. Furthermore, Maximino et al. created antimicrobial polypropylene composites utilizing copper nanoparticles functionalized with polyethyleneimine and 4-aminobutyric acid [23]. These composites were prepared using various concentrations of copper nanoparticles ranging from 0.25%, 1%, 2.5%, and 5% by weight, and their antibacterial activity toward P. aeruginosa and *S. aureus* was investigated. Notably, the 5 wt% copper–polymer nanocomposite demonstrated robust antibacterial activity compared to the other concentrations. It achieved 100% antibacterial activity within 2 h against *P. aeruginosa,* and within 4 h against *S. aureus*. Gold nanoparticles are extensively employed in various biomedical applications due to their strong stability and excellent biocompatibility [24]. They are also readily modifiable, allowing for easy customization, and their antibacterial properties can be further enhanced by altering their structure, and size, or incorporating additional ingredients. As an example, Futyra et al. developed gold–chitosan nanocomposite films and investigated their antibacterial activity against strains of *S. aureus* and *P. aeruginosa* [25]. Chitosan, a biocompatible and biodegradable polymer, was used as a reducing and stabilizing agent in the synthesis of gold nanoparticles. The resultant nanocomposite films displayed potent antibacterial properties with minimal cytotoxicity. Additionally, Zaporojtchenko et al. prepared antibacterial composite coatings composed of Ag–Au/polytetrafluorethylene (PTFE) through the co-sputtering of Ag and PTFE, where a small amount of Au (~0.1 nm) was deposited on the surface of Ag/PTFE composite films [26]. The resultant composite displayed a greater antibacterial effect than the Ag/PTFE films against *S. aureus* and *S. epidermidis*. Indeed, the concentration of nanoparticles (NPs) plays a pivotal role in toxicity, with higher concentrations leading to increased ion release. It is important to note that employing higher concentrations of metal nanoparticles might compromise the transparency of composite films.

3. Metal Oxide-Incorporated Polymer Composites

Metal oxides are substances formed when a metal reacts with oxygen. They have a wide range of properties and applications in various fields. Specifically, in the context of antimicrobial action, numerous metal oxide nanoparticles such as ZnO, CuO, MgO, SnO_2, TiO_2, and Fe_2O_3 have been investigated for their ability to counteract a broad spectrum of harmful microorganisms due to their robust durability, enduring stability, and minimal toxicity [27]. Nanocomposites directly interact with bacterial cell membranes through the electrostatic interactions between released ions and the bacterial cell wall, or the release of heavy metal ions due to surface oxidation, which causes disruption of the cell membrane, leading to bacterial damage [28][29]. Metal oxide nanoparticles, particularly those like ZnO and TiO_2, can produce reactive oxygen species (ROS) upon exposure to light or other stimuli. These ROS, such as hydrogen peroxide, singlet oxygen, superoxide anions, and hydroxyl radicals, can damage bacterial cell components like DNA, proteins, and lipids, ultimately leading to cell death [27][28]. Several studies have also demonstrated the antibacterial activity of metal oxide–polymer nanocomposite coatings in various applications [29]. Among various metal oxides, ZnO stands out as a highly promising material due to its bio-safe nature, biocompatibility, and cost-effectiveness. Its antimicrobial properties have been thoroughly investigated, showcasing effectiveness against a diverse array of pathogenic organisms [30]. For example, Dimitrakellis et al. investigated the activity of ZnO/polymethyl(methacrylate) (PMMA) nanocomposite films prepared through a solution process, both with and without atmospheric plasma treatment, against *E. coli* [31]. Plasma-treated composite films exhibited a significant enhancement in antibacterial activity due to the gradual exposure and aggregation of ZnO nanoparticles on the nanocomposite surface after plasma etching. However, nanoparticles prepared using a solution method often result in the agglomeration of the nanoparticles, which not only affects the antibacterial activity but also reduces the transmittance of the composite coatings. In contrast, metal oxide thin films prepared by sputtering exhibit excellent antibacterial activity and mechanical durability [32]. For example, ZnO-PTFE composite films prepared by the sputtering method exhibited excellent antibacterial activity and have been used for display coating applications. In addition, composite films displayed a hydrophobic nature compared with ZnO films. Copper oxide nanoparticles have also garnered significant attention for their remarkable antibacterial activity against a wide spectrum of bacteria. Haider et al. prepared Poly(lactide-co-glycolide) (PLGA)/CuO composite nanofibers by electrospinning [33]. They investigated the antimicrobial activity against *E. coli* and *S. aureus* bacterial strains, and the composite films notably inhibited the growth of both bacteria. In another report, a comparative study of the antibacterial effect of poly(butylene adipate-co-terephthalate) (PBAT)-based nanocomposites synthesized using copper nanoparticles, copper/cuprous oxide (Cu/Cu_2O) nanoparticles, and copper sulfate ($CuSO_4$) was carried out against *S. aureus*, *Acinetobacter baumanni*, *Enterococcus faecalis*, *Streptococcus mutans* [34]. Antimicrobial assessments demonstrated that the nanocomposite with Cu/Cu_2O nanoparticles resulted in antibacterial activity against *E. faecalis* and *S. mutans*, coupled and excellent bactericidal effects against *S. aureus*. Meanwhile, the composite with $CuSO_4$ exhibited effective bactericidal responses against *A. baumannii*, *E. faecalis*, and *S. mutans*, and displayed excellent efficacy against *S. aureus*. In contrast, PBAT without additives did not exhibit bactericidal properties upon contact with the bacterial strains. Furthermore, nanocomposite materials containing NiO and MgO combined with chitosan biopolymer have demonstrated antibacterial activity against both *E. coli* and *S. aureus* bacterial strains [35].

4. Carbon Derivates-Incorporated Polymer Composites

Carbon-based nanomaterials are emerging as promising platforms with diverse applications due to their unique mechanical, electronic, and biological properties. Notably, carbon nanostructures such as diamond-like carbon (DLC), graphene, graphene oxide, carbon nanotubes (CNTs), and fullerene have garnered interest attention for their potent antibacterial properties and their ability to combat a broad spectrum of pathogens [36][37]. Carbon nanomaterials exhibit an antibacterial effect through the physical disruption of cell membranes, generation of reactive oxygen species, photothermal/photocatalytic effect, inhibition of cell adhesion, electrostatic interactions, and intracellular disruption [36][38]. Indeed, recent research has highlighted the antibacterial potential of polymer composites integrated with carbon-based nanomaterials [39].

In a specific case, Santos et al. prepared an antibacterial polymer nanocomposite composed of polyvinyl N-carbazole (PVK) and graphene oxide (GO), and investigated its antibacterial properties against *E. coli* [40]. The prepared composite films exhibited improved antimicrobial activity compared with compared to both the unmodified surface and a surface modified solely with pure GO. In addition, Placha et al. investigated antibacterial activity against *S. aureus*, *E. coli*, *S. epidermidis*, and *P. aeruginosa* on functionalized graphene oxide and graphene using the quaternized statistical copolymer P(MTA90-co-DOMA10), MD10 [41]. The introduction of MD10 enhances the antibacterial effects of graphene oxide (GO) against most bacteria, except for *P. aeruginosa* and *S. aureus*. Notably, functionalized GR with MD10 exhibits the most favorable outcomes, possibly due to its increased positive charge, contributing to improved efficacy. Another study examined ultra-thin fibers manufactured from poly(methyl methacrylate) and graphene nanoplatelets (GNPs) at different concentrations (2%, 4%, and 8%) for their potential antibacterial uses against *E. coli* and *P. aeruginosa* [42]. The quantity of GNPs in composite films played a vital role in influencing bacterial growth. Surprisingly, the findings demonstrated that fibers containing 2% and 4% GNPs promoted microbial growth, while fibers with 8% GNPs displayed antimicrobial properties. Moreover, the antimicrobial activity of a polymer nanocomposite comprising 97 wt% polyvinyl-N-carbazole (PVK) and 3 wt% single-walled carbon nanotubes (SWNT) was investigated in both water suspensions, and as thin film coatings [43]. The toxic effects of different concentrations of this PVK-SWNT nanocomposite were tested against planktonic cells and biofilms of *E. coli* and *B. subtilis*. The results indicated that the PVK-SWNT nanocomposite exhibited antibacterial activity at all concentration levels. In particular, PVK-SWNT with a concentration of 1 mg/mL exhibited superior bacterial damage of 94% for *E. coli* and 90% for *B. subtilis* in planktonic cells. The antibacterial activity of several polymer composite materials against various microorganisms is summarized in **Table 1**.

Table 1. Comparison of the antibacterial activity of polymer composite materials against various microorganisms.

Composite Material	Bacteria/Virus/Fungi	Antibacterial Activity/Reduction	Ref.
Q-DMHC48/Ag NPs	***S. aureus*, *E. coli*, *P. aeruginosa*, *K. pneumoniae*, *Candida* spp., *Cryptococcus* spp.**	**>99.99% (*S. aureus*) 100% (*E. coli*)**	[16]
WPUL/Ag	***S. aureus*, *P. aeruginosa***	**99.99%, 99.99%,**	[19]
PBAT/Cu, PBAT/Cu|Cu_2O, PBAT/$CuSO_4$	***S. aureus*, *A. baumanni*, *E. faecalis*, *S. mutan***	**-**	[34]
Cellulose/Cu nanofillers	***S. aureus*, *K. pneumoniae***	**-**	[21]
Polyurethane/Cu	***S. aureus*, *E. coli***	**2 Log10 < cell density < 3 Log10, ≥3 Log10**	[22]
Polypropylene/Cu NPs	***S. aureus*, *P. aeruginosa***	**100%**	[23]
Chitosan/Au NPs	***S. aureus*, *P. aeruginosa***	**-**	[25]
Ag-Au/PTFE	***S. aureus*, *S. epidermidis***	**-**	[26]
PMMA/ZnO	***E. coli***	**1 Log CFU/ml**	[31]
PLGA/CuO NFs	***S. aureus*, *E. coli***	**-**	[33]
Chitosan/NiO-MgO	***S. aureus*, *E. coli***	**98%, 92.3%**	[35]
Cationic Polymers-GO and GR	***S. aureus*, *E. coli*, *S. epidermidis*, *P. aeruginosa***	**-**	[41]
Polyvinyl N-carbazole/SWNT	***E. coli*, *B. subtilis***	**94%, 90%**	[43]
Polyvinyl N-carbazole/GO	***E. coli***	**90%**	[40]

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