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Review

Antimicrobial Nanomaterials: A Review

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Abstract: Microbial colonization on various surfaces is a serious problem. Biofilms from these microbes pose serious health and economic threats. In addition, the recent global pandemic has also attracted great interest in the latest techniques and technology for antimicrobial surface coatings. Incorporating antimicrobial nanocompounds into materials to prevent microbial adhesion or kill microorganisms has become an increasingly challenging strategy. Recently, many studies have been conducted on the preparation of nanomaterials with antimicrobial properties against diseases caused by pathogens. Despite tremendous efforts to produce antibacterial materials, there is little systematic research on antimicrobial coatings. In this article, we set out to provide a comprehensive overview of nanomaterials-based antimicrobial coatings that can be used to stop the spread of contamination to surfaces. Typically, surfaces can be simple deposits of nanomaterials, embedded nanomaterials, as well as nanotubes, nanowires, nanocolumns, nanofibers, nanoneedles, and bio-inspired structures.

Keywords: antibacterial; nanomaterial; gold; silver; titanium dioxide; zinc oxide; copper oxide

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1. Introduction

The developing technology in the field of antimicrobial coating can make an important contribution to overcoming biofilms in various fields, such as food, medicine, and agriculture. Biofilm-associated infections cause serious public health burdens and major economic losses [1]. Pathogenic infectious agents that form biofilms remain alive on surfaces for a long time. It is also known that multi-drug-resistant bacterial strains can survive for weeks on different surfaces in the hospital [2]. The National Institute of Health announced that biofilms are responsible for 80% of the total number of microbial infections in humans [3], including meningitis, cystic fibrosis, kidney infections, endocarditis, rhinosinusitis, periodontitis, non-healing chronic wounds, osteomyelitis, prosthetic, and implantable medical device infections [4].

The main difficulties in the treatment of biofilms are that they are difficult to remove in the clinic and are not easy to diagnose due to their high tolerance to antibiotics [5]. Numerous molecules that inhibit biofilm formation or disperse the resulting biofilms have been identified. Nanoparticles, antibiotics, antimicrobial peptides, enzymes, quaternary ammonium compounds, superhydrophobic coatings, and anti-adhesive polymers are used as antimicrobial strategy types in surface coatings [6–8]. With the latest developments in nanotechnology, new opportunities for effective biofilm treatment and control have become the focus of attention. Nanotechnology-based strategies allow the fabrication of nanoscale surfaces that can both reduce bacterial adhesion and increase osseointegration without the use of biomolecules or antibiotics, as well as the promise of biomaterials and medical devices to prevent drug-resistant biofilm infections [9]. Because NPs are very small in size, they cause irreversible damage to DNA and cell membranes by penetrating microbial cell walls and even biofilm layers. In addition, these structures have long plasma half-lives and facilitate drug loading and the targeting of entities due to their high surface-to-volume ratio [10]. The antibacterial activity of most metal-based coatings is attributed to their

oligodynamic effect, which is ionic or nanoform dependent, as opposed to bulk properties. The use of metals such as gold, zinc, silver, and copper for various antimicrobial purposes has been documented since ancient times. These substances are antimicrobial agents that have the power to kill Gram-negative and Gram-positive bacteria, viruses, protozoa, and fungi. Therefore, these substances have been used in antimicrobial-based products and the field of medicine for a long time [11,12]. Antimicrobial surfaces should aim to prevent bacteria colonization and attachment [13]. Different strategies can limit or prevent bacterial colonization on the biomaterial surface. However, there are still concerns regarding the evolution of antimicrobial resistance, and the risks of toxicity for these surfaces are high.

In addition, biocidal agents that deplete over time also lose their bioactivity [14]. All these limitations highlight the need for new antimicrobial surface coatings. The development of nanotechnology and biomaterials has led to antimicrobial nanoparticles becoming promising candidates in a variety of applications. In addition, with the developments in nanotechnology, the ability of biomaterials coated with antimicrobial nanoparticles to combat microbial adhesion, vitality, and biofilm formation with versatile antimicrobial mechanisms is promising for new-generation implants. However, despite these, there are many difficulties in applying nanoparticles to implant surfaces while preserving their antimicrobial properties [15]. Here, we review the potential applications of the designed antimicrobial nanomaterial coatings and the challenges associated with them.

2. Classification of Antimicrobial Nanomaterials

Antibiotic resistance is recognized worldwide as one of the greatest threats to public health. It has been noted that more than 70% of infections caused by bacteria can develop resistance to the main antimicrobial agents used in clinical practice. It has also been determined that approximately 79% of bacteria develop resistance to one or more antibiotics [16]. The effects of antibiotic resistance are not only limited to increased mortality and health complication risks but also lead to increased healthcare costs. The development of new antibiotics is an effective solution to this antibiotic resistance problem. However, the development of new antibiotics often takes years, making it impossible to quickly reduce the immediate problem of antibacterial resistance. In addition, this process is costly, and new antibiotics will only be effective for a limited time until resistance reappears. This has economically hindered the development of new antibiotic classes [17,18]. Therefore, there is a great need to develop a potent antimicrobial agent. Nanoparticles have emerged as new tools that can be used against bacterial infections to help overcome antibiotic resistance and the barriers faced by conventional antimicrobials [19]. Metals, including copper, silver, and zinc, have been used in modern medicine for centuries for infection control due to their antibacterial properties. Nanomaterials containing these metals can be found in various forms, such as nanoparticles of metal or metal oxides and composite materials with different metal layers [20]. With advances in nanotechnology, functional nanomaterials with unique chemical and physical properties have been developed. Nanoparticles with large surface area-to-volume ratio properties offer numerous options for developing agents to treat microbial infections. Metal and metal oxide NPs from this class are very promising candidates as antimicrobial agents [21]. Nanoparticles are materials that show antimicrobial and biocidal activity against bacteria, fungi, and viruses. Nanomaterials derived from gold, silver, titanium dioxide, zinc oxide, and copper oxide are widely used in various fields such as cosmetics, device coatings, and food preservation [22]. In Figure 1a,b cell wall structure of gram-negative and gram-positive bacterias and antimicrobial mechanisms of metal ions is shown respectively [23].

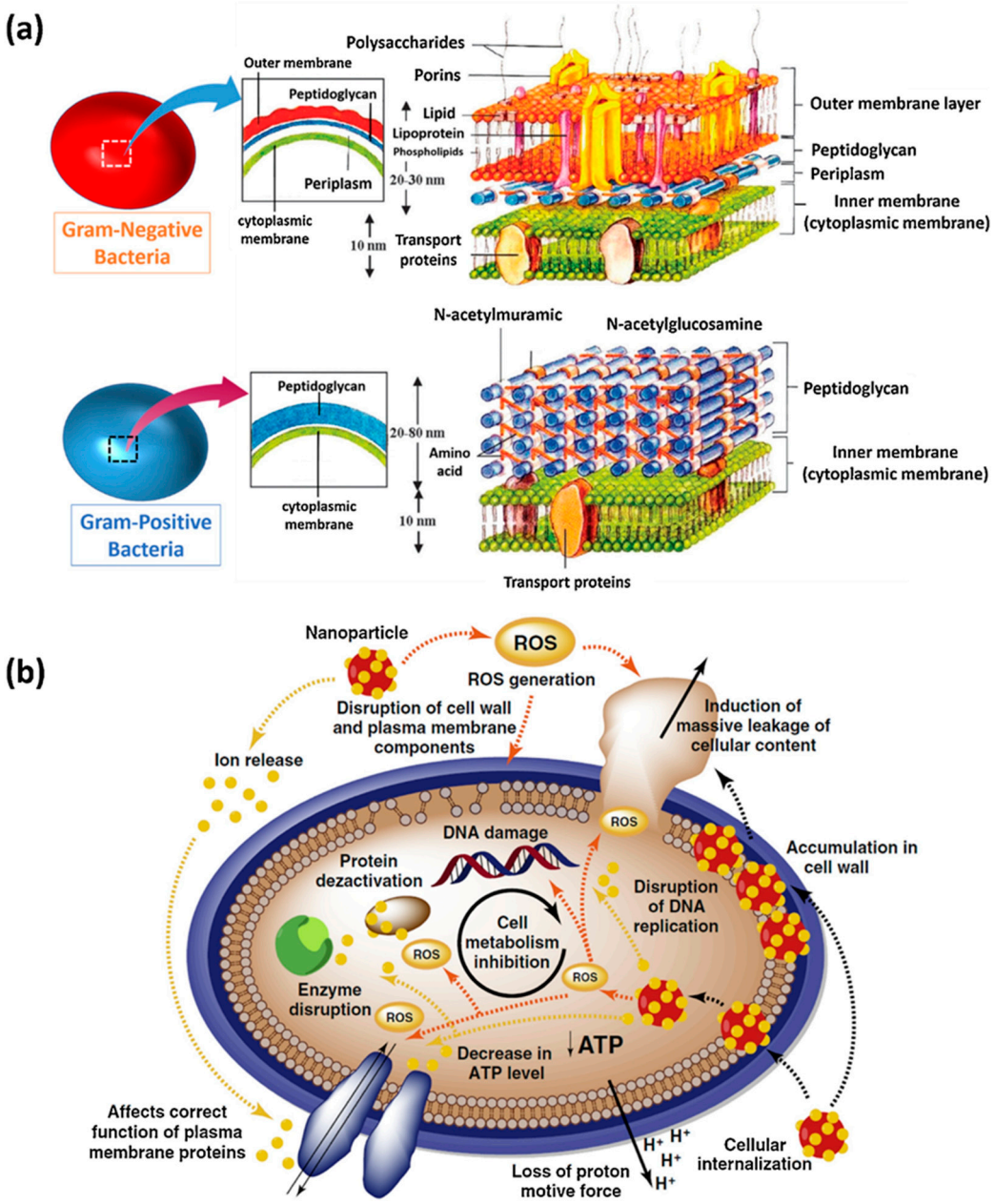

Figure 1. (**a**) Comparison between Gram-negative and Gram-positive bacteria cell wall structures and (**b**) schematic representation of antimicrobial mechanisms of metal ions (shown with permission from the authors of [23]).

2.1. Silver-Based Nanomaterials

Silver-based nanomaterials in different forms such as particles, plates, and wires are used as components in various products and applications [24]. Silver nanoparticles are one of the most studied nanomaterials due to their wide range of applications. These materials are of great interest due to their strong antimicrobial properties against bacteria, viruses, and fungi. In addition, silver nanoparticles (AgNPs) can be applied as disinfectants and exhibit synergistic effects with antibiotics [25].

Silver nanoparticles are used as antibacterial agents in cosmetics, healthcare products, textile fabrics, dressings, and coatings, as well as in clinical applications, e.g., for the treatment of chronic ulcers, antibiotic-resistant diabetic wounds, and burns. In addition, when AgNPs are used in wound therapy, they elicit abundant collagen deposition, which can accelerate wound healing and exhibit anti-inflammatory effects [26]. Silver nanomaterials

(especially those with dimensions $\leq$ 10 nm) are toxic to many human cell lines and cause cytotoxicity depending on factors such as size, time, and dose. To solve this problem, the immobilization of these structures on various support materials such as polymers, activated carbon, metal oxides, and graphene oxide has been investigated. The modification of silver nanoparticles with titanate nanotubes changes their physicochemical properties (such as stability, size, oxidation state, and shape), resulting in enhanced antibacterial, catalytic, and photocatalytic activity [27].

2.2. Gold-Based Nanomaterials

Gold-based nanomaterials can be engineered in various ways to ensure antimicrobial activity. Gold-containing nanomaterials include nanorods, nanolatches, nanoclusters of nanorods, and nanoshells. Optimizing the properties of these substances can be achieved by conjugation with other compounds or by modifying their nanostructure. Antimicrobial activity can also be achieved by the conjugation of gold nanoparticles with antibodies and different antimicrobial agents. Antibiotic-conjugated gold nanoparticles show potent antimicrobial activity against various bacteria and antibiotic-resistant strains [28]. Various metal nanoclusters, such as silver, copper, and gold, are used as antibacterial agents. Gold nanoclusters (AuNCs) from metal nanoclusters have exhibited superior properties in imaging, detection, and biomedical applications. In therapy, AuNCs conjugated with different surface ligands have been widely applied as antimicrobial agents due to their easy modification, polyvalent effects, photothermal stability, and high biocompatibility [29]. Gold nanoparticles have been developed as strong candidates due to their ease of surface functionalization and high biocompatibility [30]. In pharmacology, AuNPs offer anti-angiogenesis, anti-HIV, anti-malarial, antimicrobial, and anti-arthritic activity. Biomedical uses of these materials include gene therapy, drug delivery, diagnostics, and catalysts for medical therapy [31].

2.3. Titanium Dioxide (TiO_2) Nanomaterials

The important discovery of ultraviolet (UV) light-mediated water separation on the titanium dioxide (TiO_2) surface and various applications of TiO_2 nanoparticles, especially nanomedicine and nanobiotechnology, have been widely studied [32]. Titanium dioxide is highly attractive for photocatalytic bactericidal activity compared to other nanoparticles due to its chemical stability, natural abundance, and relatively low cost [33]. In addition, TiO_2 nanoparticles are also widely preferred in biological and environmental remediation applications due to their good thermal stability, chemical biocompatibility, unique photocatalytic activity and physicochemical properties, non-toxicity, and high surface-area-to-volume ratio [34]. TiO_2 nanoparticles have broad activity against fungi and bacterial microorganisms, and these properties are of great interest for multidrug-resistant strains. In addition, due to the non-contact biocidal effect and environmental friendliness of TiO_2-based nanocomposites, it is not necessary to release potentially toxic nanoparticles into the environment to achieve disinfection properties [35].

2.4. Zinc and Zinc Oxide Based-Nanomaterials

The use of inorganic antimicrobial agents against infections offers advantages such as good selectivity, lower toxicity, lower microbial resistance, and higher stability compared to organic antimicrobials. Among them, zinc oxide (ZnO) offers advantages such as high heat resistance, higher selectivity, lower cytotoxicity, and higher stability compared to inorganic antimicrobials. When it comes to antimicrobial application, NPs exhibit superior properties compared to bulk materials. It can improve interactions with cells and optimize antimicrobial activity due to increased contact surface area [36]. Various studies have shown that smaller ZnO NPs have better antimicrobial activity compared to larger nanoparticles [37,38]. The application of ZnO in healthcare products has attracted particular attention because of its UV radiation-blocking capability. It has been supported by various studies that ZnO NPs have antibacterial activity and that these structures have high efficacy

against different bacteria [39]. In medicine and biology, the antimicrobial and fungicidal activities of ZnO-NPs, their cytostatic activity against cancer cells, their anti-inflammatory activity, their use in bioimaging due to their chemiluminescent properties, their ability to accelerate wound healing, and their antidiabetic properties are of great interest [40].

2.5. Copper and Copper Oxide-Based Nanomaterials

Copper and copper oxide-based nanomaterials are very interesting due to their unique properties that enable them to be used in many fields such as sensors, optics, solar cells, catalysts, electronics, remediation applications, and antimicrobials [41]. Copper is a highly conductive material and is cheaper than materials such as gold (Au) and silver (Ag). Since CuO phases are more stable thermodynamically than pure copper, most of the synthesized copper nanoparticles (Cu NPs) have surface oxide layers. The CuO nanostructure is also a p-type semiconductor with a monoclinic structure and high dielectric constant [42]. The generally accepted antibacterial action mechanism of copper-based nanomaterials is based on the release of Cu^{2+} ions. Copper ions can damage the bacterial cell membrane and enter cells to disrupt enzyme function, causing bacterial death [43]. Copper nanoparticles are highly reactive antimicrobial materials due to their high surface-to-volume ratio. Among metal oxide nanoparticles, CuO NPs are of great importance as they are the simplest member of the copper group. CuO NPs are structured with important antimicrobial properties that inhibit the growth of viruses, fungi, bacteria, and algae [44]. Copper-based compounds are materials with highly effective biocidal properties that are often used in pesticide formulations and various applications in the health field [45]. In addition, CuO NPs are widely used for different applications due to their unique properties, optical applications, gas sensors, solar cells, high-temperature superconductors, lubricants, catalytic applications, and medical applications [46].

3. Applications of Antimicrobial Nanomaterials

One factor that is particularly important in the spread of infectious diseases is the ability of bacteria to survive on surfaces, both in healthcare facilities and on everyday surfaces. Inadequate hygiene practices can result in the growth of microbial sanitizer resistance, the occurrence of foodborne illnesses, and financial losses. Food deterioration, biofilm development, biofouling, microbially influenced corrosion, and outbreaks of foodborne illness are just a few of the problems that microbial resistance can pose for the food industry [47]. Therefore, the prevalence of foodborne illnesses linked to bacterial resistance is an urgent public health issue that requires careful monitoring [48].

Infectious diseases caused by expected and unexpected viruses and bacteria are largely treated via the use of nanotechnology. Generally, antimicrobial agents are chemical compounds that are either bioactive polymers or synthesized polymers, either alone or in combination with nanoparticles (NPs) [49]. Due to their high surface-to-volume ratio and well-established antibacterial capabilities, NPs in particular exhibit a substantial reaction even at low concentrations [50]. Their primary purpose is to act as an antimicrobial agent, preventing and killing pathogenic viruses, fungi, and bacteria. Many organic substances, including polymers and biopolymers, have recently demonstrated promise as antiviral and antibacterial medicines to treat diseases brought on by dangerous bacteria and viruses [51].

Biocidal coatings are formed using passive pathogenic repulsive surfaces and active antimicrobials, micro- and nanostructuring, nanomaterials, and chemical modifications [52]. Singh et al. emphasized the importance of the latest methods for the production of nanomaterials, such as nanoparticles with green synthesis [53,54]. The green synthesis of metal and metal oxide nanoparticles using components of various plants for anticancer and antibacterial properties was supported by Bukhari et al. [55].

Strategies for improving nanoparticles' antibacterial capability through surface modification and their potential as a delivery system for antibiotics have also been investigated. Barabadi et al. aimed to fabricate silver nanoparticles (AgNPs) obtained from the Zataria multiflora and compare their antibacterial and biofilm-inhibitory properties to those of

commercially produced AgNPs (C-AgNPs) against *Staphylococcus aureus* ATCC 25923 [56]. Figure 2 shows images of biosynthesized AgNPs, characterization of AgNPs, and also the biofilm inhibitory and antimicrobial activity of AgNPs.

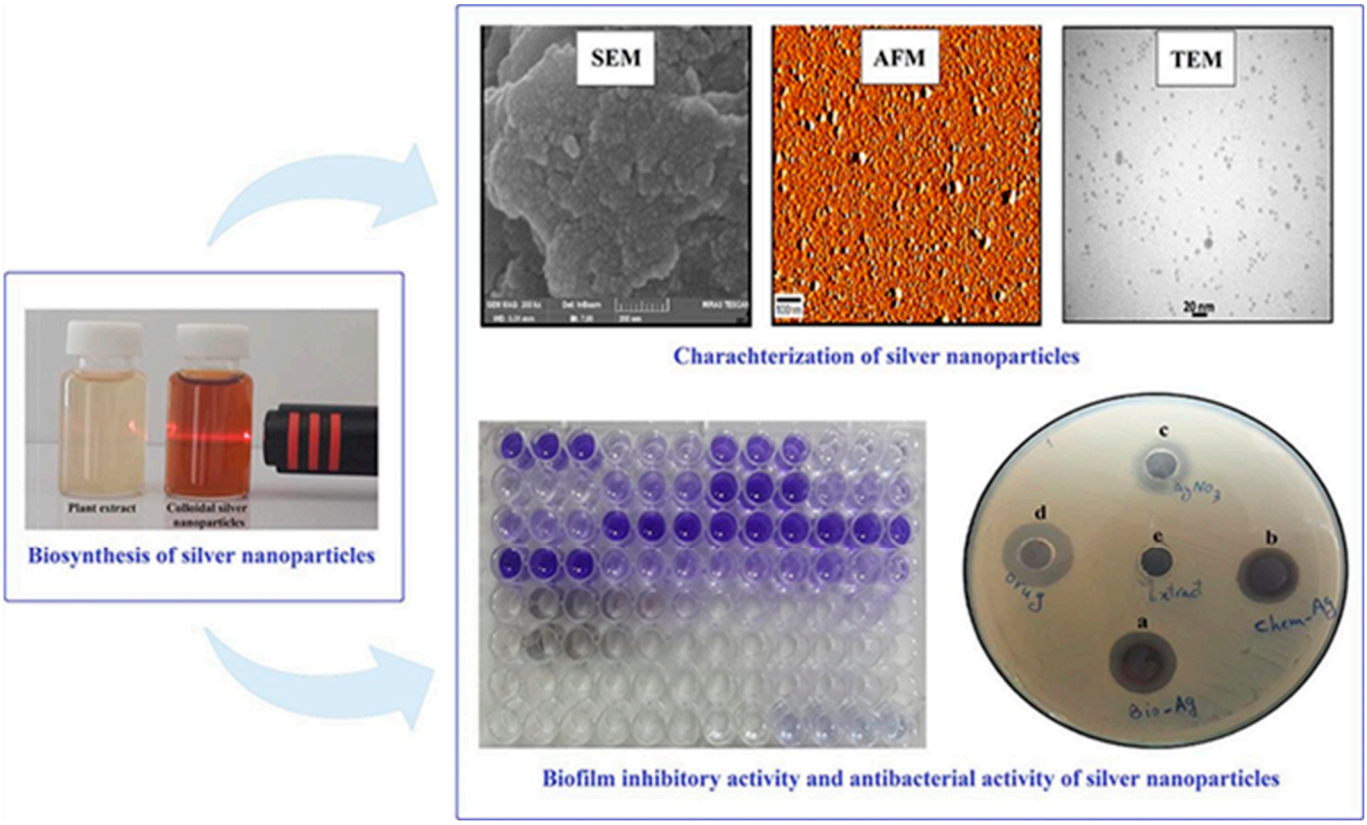

Figure 2. Biosynthesis, characterization, and antibacterial activity of AgNPs. (with permission from the authors of [56]).

The study of nanotechnology has attracted interest in several disciplines, including food, medicine, cosmetics, lubricants, agriculture, fuel additives, paints, and others. Recent developments in green nanoparticle synthesis for the treatment of wounds and bacteria were reviewed by Nandhini [57].

Since biosynthesis does not use toxic chemicals and is eco-friendly, it is favored among other methods of creating nanoparticles. Plant, fungal, algal, and cyanobacterial extract solutions have been employed as nucleation/capping agents in the green bioprocess to produce effective nanomaterials for cutting-edge medical applications. Synthesized nanoparticles have shown wound-healing antibacterial, anticancer, and antifungal properties, as shown in Figure 3. Rizki et al. evaluated the use of marine organisms for the environmentally friendly synthesis of gold and silver nanoparticles and their applications [58].

Wide-spectrum drugs are frequently the target of antimicrobial resistance (AMR). Nanomaterials are being used in some applications, including antimicrobial coatings, to fight AMR, which is a burgeoning area of research. Although their modes of action are different, metal and metal oxide nanoparticles both seem to be efficient in that regard.

The presence of bacterial strains that are resistant to one or more medications may lead to wound infections. As a result, NPs with antibacterial activity that are present in medical dressings and plasters are a fantastic substitute for betadine, alcohol, and antibiotics. Silver (Ag) is being increasingly used as an antibacterial agent. With outstanding anti-inflammatory results, AgNPs have been applied in a number of methods to treat wounds [59]. AgNPs can build up in the human body, are highly expensive, challenging to obtain, and frequently unstable in a range of settings [60].

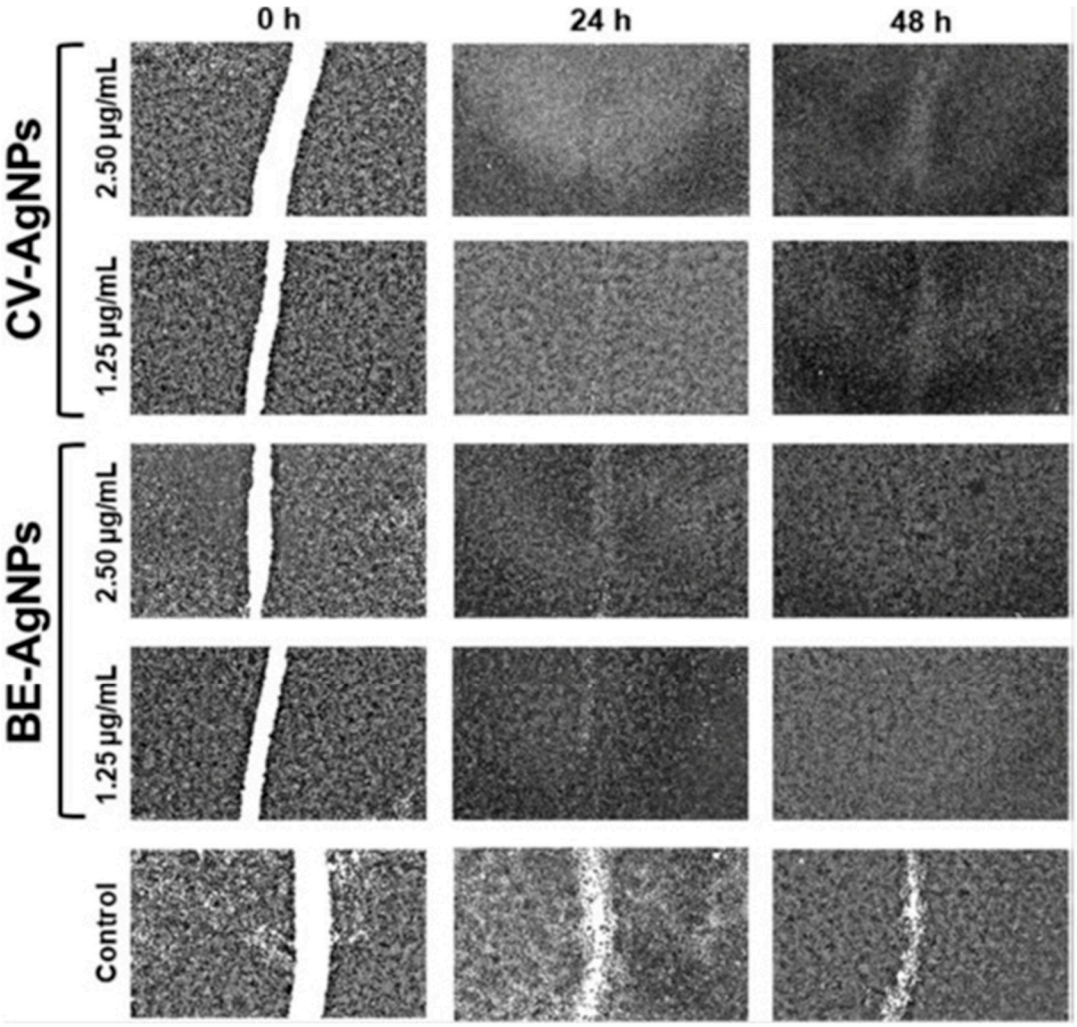

Figure 3. In vitro wound healing effectiveness of CV-AgNPs and BE-AgNPs at 2.50 g/mL and 1.25 g/mL for 24 and 48 h in L929 cell line (with permission from the authors [57]).

Since ZnO and TiO_2 have no impact on human cells, NP-based wound care solutions utilizing these two oxides are relatively new compared to conventional materials and are safe to employ as antibacterial agents. Veselova et al. investigated the long-term antibacterial efficacy of textiles coated with ZnO and TiO_2 nanoparticles in a tropical environment [61]. Cotton bandages containing Ag- and ZnO-NPs and mixed Ag/ZnO-NPs show antibacterial activity and high resistance to *Acinetobacter baumannii* and *Pseudomonas aeruginosa* [62]. Recent advances in nanotechnology have led to the creation of copper nanoparticles (CuNPs) with enhanced antibacterial properties. The effectiveness and mechanism of the antimicrobial activity of copper against microorganisms, which has been used as an antimicrobial agent from ancient civilizations to the present, has been determined. Overall, copper has an inherent ability to fight against fungi, viruses, and bacteria. The size and concentration of CuNPs appear to have a greater impact on their activity; the smaller and more efficient the nanoparticles, the more effect they have [63]. Small-size nanoparticles find it easier to penetrate cells. This suggests that the plasma membrane is weakened by the CuNPs' interactions with the cell wall. Recent advances in the use of copper as an antimicrobial agent were reviewed by Salah et al. [64]. The main method by which copper nanoparticles cause the death of many bacteria is depicted in Figure 4.

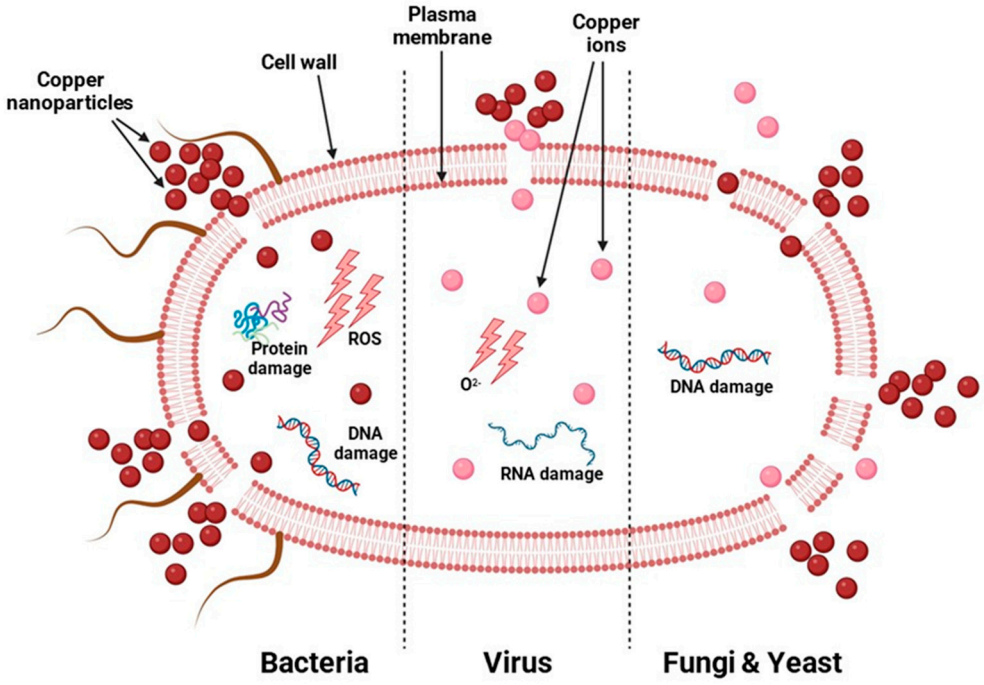

Figure 4. Copper nanoparticles are the main cause of death in several bacteria. (Adapted from [64]).

Due to their extremely small sizes, high surface area/mass ratio, and special physical and chemical properties, antimicrobial nanoparticles seem promising. Many studies have been conducted on the potential of metal nanoparticles as antibacterial agents, and they are currently viewed as an alternate strategy to deal with the problem of bacterial multidrug resistance. Because pathogens (disease-causing microbes) may persist on surfaces for up to a few days and spread through them often, surfaces become a major site of pathogen transmission. Song and Ge discussed the creation, uses, and underlying mechanisms of antibacterial nanoparticles in the fields of periodontics, endodontics, orthodontics, implantology, restorative dentistry, and implantology [65].

Innovative nanoparticles for medical applications were created using a variety of metal salts, including silver, zinc, titanium, and other inorganic salts. The use of inorganic nanoparticles to fight fungal infections in the antimicrobial-resistant era was reviewed by Huang et al. [66]. Sharma et al. created antimicrobial polymeric surfaces with incorporated silver nanoparticles [67]. In addition, zinc oxide nanoparticles are widely used in dermatology due to their antibacterial activity and high safety profile. Dacrory et al. discovered that the antifungal activity of DAC/Gly/Zn was superior to clotrimazole when used as a conventional antifungal against *A. niger* and *A. fumigatus*, as demonstrated in Figure 5 [68].

The application of ZnO, TiO_2, and Zn/TiO_x nanoparticles produced by plasma for the creation of antibacterial wound-healing viscose patches was carried out by Pirotkin et al. In their study, underwater pulse discharge plasma was used to synthesize zinc oxide (ZnO) nanoparticles (NPs), TiO_2 NPs, and mixed Zn/TiO_x-NPs in an environmentally friendly, energy-efficient, and practical manner [59].

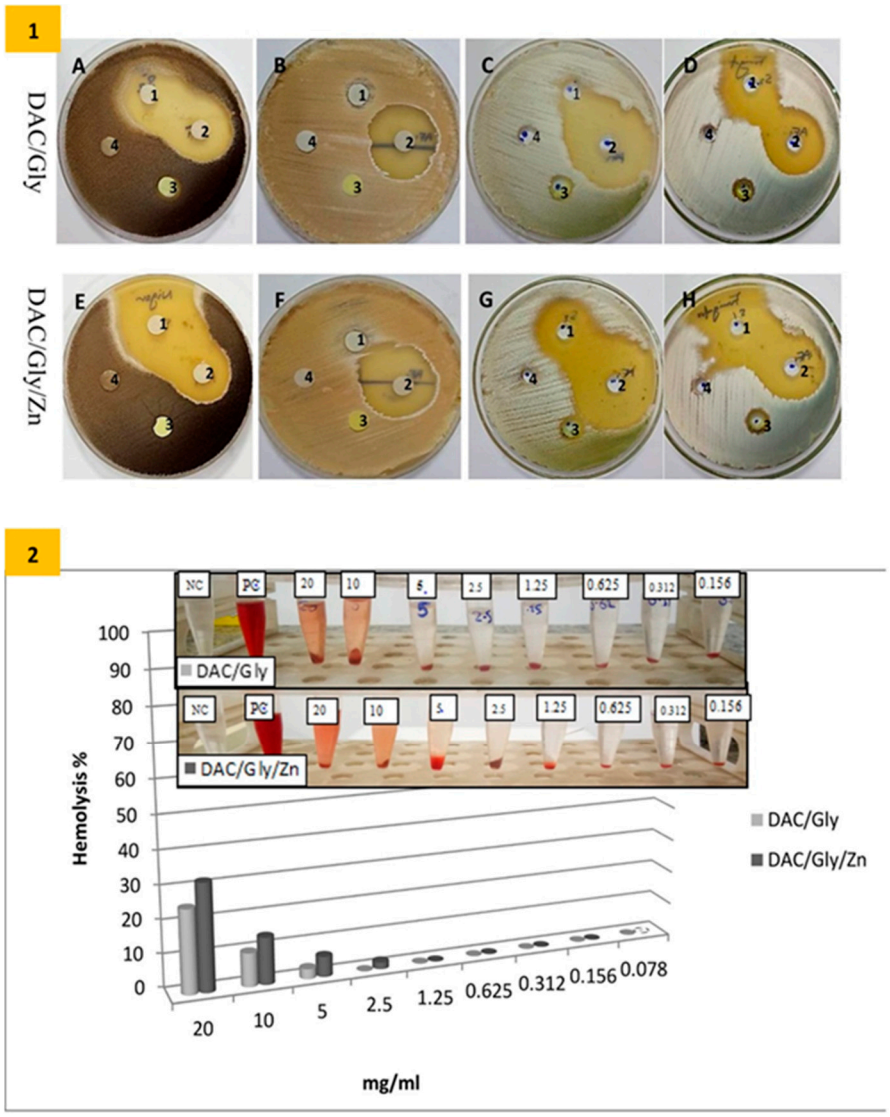

Figure 5. (1) Antifungal activity of DAC/Gly (**A**–**D**) and DAC/Gly/Zn (**E**–**H**): *A. niger* (**A**,**E**), *A. terreus* (**B**,**F**), *A. flavus* (**C**,**G**), *A. funigatus* (**D**,**H**). Numbers in the top row (1–4) pertain to DAC/Gly, clotrimazole, nystatin, and DMSO, respectively; numbers in the bottom row pertain to DAC/Gly/Zn, clotrimazole, nystatin, and DMSO, respectively. (2) Haemcompatbility of the DAC/Gly and DAC/Gly/Zn. NC: Negative control; PC: Positive control. (With permission from the authors of [68]).

Depending on the type of genome they possess, viruses either have double-stranded or single-stranded nucleic acids. Since RNA viruses evolve and mutate more quickly than other types of viruses, they are more likely to spread illnesses in clusters. Severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2), responsible for the COVID-19 pandemic, is a highly infectious virus. Influenza A, which causes the common flu, can lead to pneumonia and high fever. The norovirus, sometimes referred to as the "winter vomiting bug", is what causes widespread gastroenteritis. Therefore, the need for strategies to limit the spread of this highly contagious virus is great. Hodek et al. examined the usage of silver, zinc, and copper as components in a sol-gel hybrid coating [69]. HIV-1, herpes simplex virus (HSV), dengue, influenza, and coxsackie viruses were utilized in antiviral tests to provide a thorough analysis of enveloped and non-enveloped and DNA- and RNA-based viruses. Hasan et al. recently examined nanostructured surfaces to determine whether they have antiviral properties. It was discovered that rhinovirus-16 (RV-16) was extremely vulnerable to a loss of viability for 24 h [70]. Chitosan/poly(4-vinyl pyridine) (chit/P4VP) coatings

were electrophoretically deposited (EPD) on titanium substrates that had previously been coated with silver nanoparticles (AgNPs) at various P4VP suspension concentrations and voltage levels [71]. Antiviral coatings over surfaces used for daily handling are in high demand all around the world as they can help prevent viral contamination. Universal antiviral nanocoating-based surfaces were reviewed by Vijayan and coworkers [72]. Photocatalytic TiO_2 nanomaterials as potential antiviral and antimicrobial agents were discussed by Prakash et al. [73]. Recent breakthroughs in nanostructured antiviral coating and filtration materials were also discussed in a mini review by Dahanayake et al. [74]. He et al. examined the antiviral properties of silver nanoparticles against SARS-CoV-2 virus [75]. Tavakoli and Hashemzadeh have shown that CuO-NPs have an antiviral effect for HSV-1 inactivation via the oxidation of viral proteins or degradation of the viral genome [76]. In another study, Jana et al. produced polysaccharide-encapsulated and in situ nanostructured ZnO NPs with antiviral potential and low cytotoxicity (viability ~90%) by simply controlling the formation within spatial 3D networks of hydrogels. These compounds, ChB@ZnO and ChH@ZnO, showed highly potent antiviral activity against Human cytomegalovirus (HCMV) [77].

Various carbon-based nanomaterials, such as carbon nanotubes, graphene, carbon dots, and nanotubes, have properties including excellent surface chemical structures, biocompatibility, low toxicity, and easy functionalization. Carbon-based nanomaterials are promising as they could have inhibitory effects against pathogenic viruses [78]. rGO-FET was produced for the sensitive and selective detection of one of the most common viruses, the sexually transmitted human papillomavirus (HPV). The sensitive and selective detection of the HPV-16 E7 protein is based on the attractive semiconductor properties of pyrene-modified Rgo [79]. Donskyi et al. investigated graphene platforms with dual sulfate/alkyl functions for the inhibition of SARS-CoV-2. In this study, graphene derivatives with long alkyl chains (>C9) indicated that they inhibited coronavirus replication by disrupting the viral envelope. Graphene platforms were found to exhibit potent antiviral activity against native SARS-CoV-2 without showing significant toxicity to human cells [80].

Dendrimers are attractive carriers for the delivery of antiviral agents because of their nanosize and structural homogeneity. The antiviral activity of dendrimers is attributed to their multivalence, symmetrical, monodisperse molecular structure, and the presence of multiple functional groups on the surface [81]. Asgary et al. investigated the adjuvant effect of the G2 dendrimer in the rabies vaccine. In this study, a nonlinear spherical G2 dendrimer containing polyethylene glycol 600 (PEG-600) and citric acid was synthesized. The toxicity of the dendrimer was studied in vitro on the J774A.1 cell line. Also in this study, the adjuvant effect of the G2 dendrimer in the elevation of neutralizing antibodies to the rabies virus was demonstrated for the first time [82]. A study performed by Ailincai et al. reported the synthesis and characterization of the drug delivery system (DDS) for the co-administration of antiviral and antifungal agents. Based on the reversible imine linkages between chitosan oligomers and 2-formiphenyl boronic acid, the encapsulation of tenofovir in hydrogels synthesized DDSs that can consistently co-deliver both the antimicrobial aldehyde and the antiviral drug (Figure 6). The resulting DDSs have been shown to offer hemocompatibility, biodegradability, and in vivo biocompatibility, along with an ability to co-release the drug and antifungal aldehyde, making them promising materials for the treatment of HIV infection and associated co-infection symptoms [83].

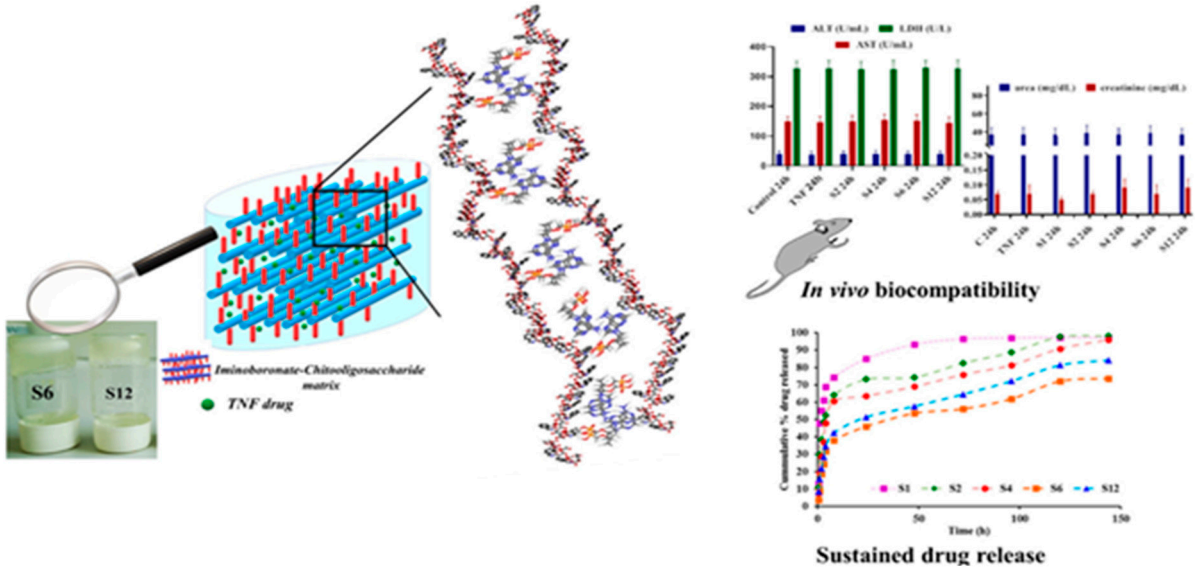

Figure 6. Synthetic pathway for the obtaining of DDSs (with permission from the authors of [83]).

Yoon et al. prepared polyacrylonitrile (PAN) nanofibers containing various copper salts and ZnO nanorods via electrospinning and investigated the antiviral activities of these structures. The antiviral activities of the synthesized NFs were evaluated by the inactivation of bacteriophage φx174 under both visible light irradiation and dark conditions (Figure 7). In this study, CuBr/ZnO NFs were found to be more effective than NFs containing other copper compounds, and the physical contact of the virus with Cu(I) ions containing CuBr well dispersed in NFs was shown to be the main cause of virus inactivation [84].

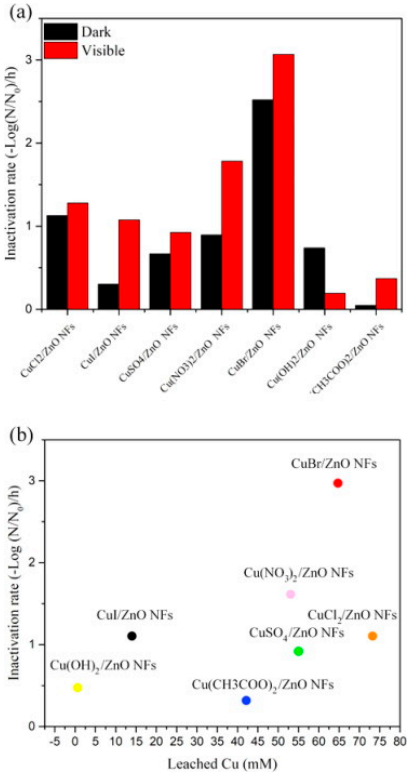

Figure 7. (**a**) Inactivation rate of produced nanofibers under dark and visible light irradiation conditions and (**b**) amount of leached ions during the antiviral test (with permission from the authors of [84]).

Attia et al. used a green synthesis approach to develop renewable, innovative, efficient, and cost-effective antibacterial and antivirus coatings for textile fabrics. In this study, rennet casein solution was prepared from cow's milk source as a biopolymer and halloysite nanotubes (HNTs) were uniformly dispersed, producing one-dimensional new nanocomposites. Nanocomposites containing different HNT mass charges (10%, 30% and 50%) were coated on different fabrics, and the toxic gas suppression, flammability, reinforcement, and antiviral and antibacterial properties of coated textile fabrics were significantly improved. In addition, it has been determined that the developed coating provides more than 90% antiviral inhibition for adenovirus and herpesvirus. The morphological collapse of adenovirus- and herpesvirus-infected cells was demonstrated (by the arrows) on the surface of the different coatings produced (Figures 8 and 9) [85].

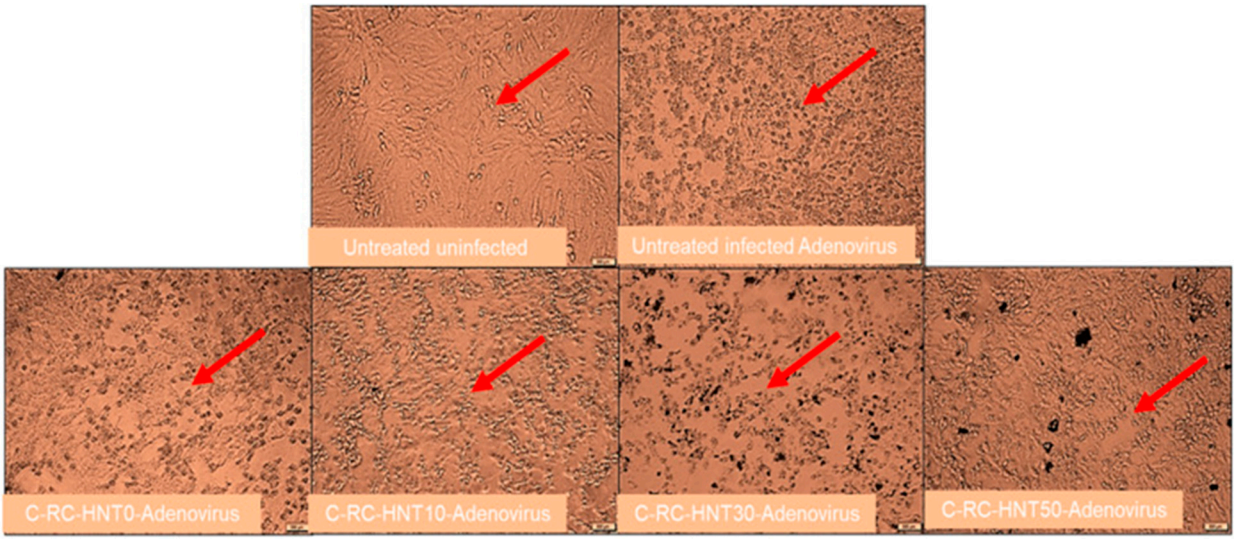

Figure 8. Morphological changes in the produced coatings with adenovirus infected/non-infected healthy cells (with permission from the authors of [85]).

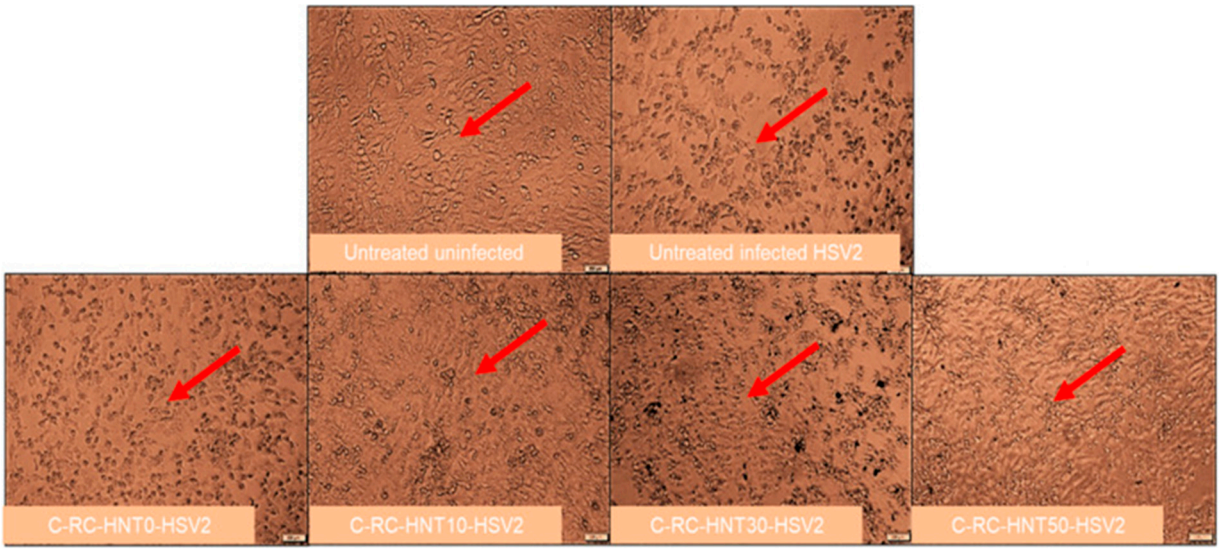

Figure 9. Morphological changes in the produced coatings with herpesvirus-infected/non-infected healthy cells (with permission from the authors of [85]).

Lin et al. developed quercetin-derived carbonized nanogels (CNGsQur) that exhibit antioxidant, potent viral inhibitor, and anti-inflammatory activities to prevent the spread of IAVs (Influenza A viruses). It has been determined that the developed CNGQur270 can

suppress inflammation via scavenging free radicals and the formation of reactive oxygen species (ROS), and the superior biocompatibility of CNGsQur270 has been demonstrated in in vivo and in vitro cytotoxicity experiments. The antiviral properties of CNGsQur270 have also been found to correlate with their adhesion to the virus envelope, as evidenced by their effective inhibition of H1N1 adhesion to the cell membrane of MDCK cells. In addition, CNGsQur270 has been reported to have a much higher selectivity index (SI > 857.1) compared to antiviral drugs in current clinical use. For these reasons, CNGsQur270 has been shown to have great potential as a viable alternative for the treatment of H1N1. Synthesis of carbonized nanogels (CNGs) from quercetin in one step and their applications associated with the treatment of H1N1 virus were shown in Figure 10A,B respectively [86].

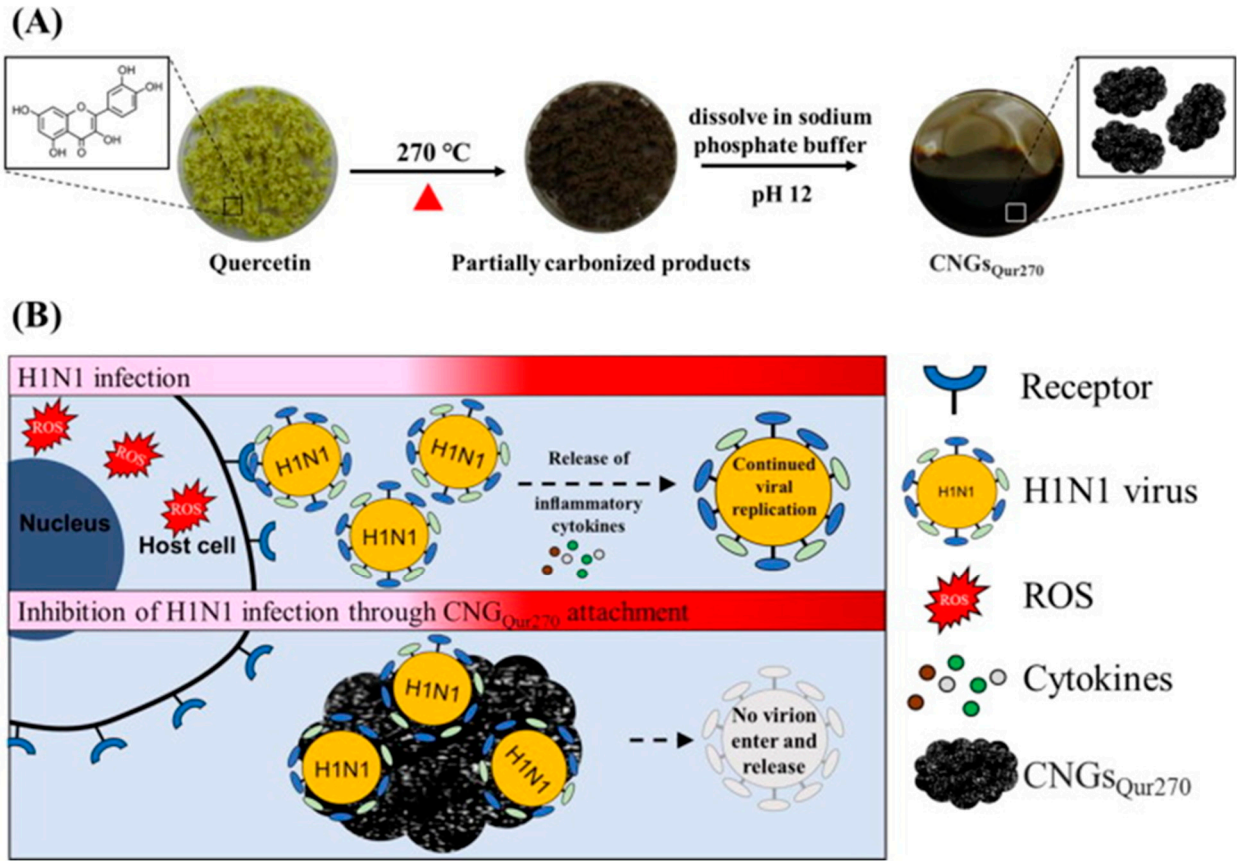

Figure 10. (**A**) Synthesis of carbonized nanogels (CNGs) from quercetin in one step and (**B**) their applications associated with the treatment of H1N1 virus through blocking viral binding to lung epithelial cells, alleviating oxidative stress and inflammation (with permission from the authors of [86]).

The long-acting injectability (LAI) of oleogels loaded with Tenofovir alafenamide-chitosan polymeric nanoparticles developed with ethyl cellulose and sesame oil for the prolonged release of the drug was reported for the first time by Narayanan et al. It has been shown to be a unique long-acting parenteral formulation for chronic anti-retroviral therapy by ex vivo and in vitro studies. The cell uptake of nanoparticles was evaluated using L929 cell lines (Figure 11), and the ability of chitosan nanoparticles to enter cells was confirmed. In addition, oleogels have been shown to exhibit prolonged drug release (56%) over 16 days, resulting in a 10-fold reduction in drug permeability. Overall, it has been reported that the developed Tenofovir alafenamide-chitosan polymeric nanoparticles loaded with ethylcellulose oleogels could potentially be used as an effective injectable system for the treatment of patients infected with HIV/AIDS [87].

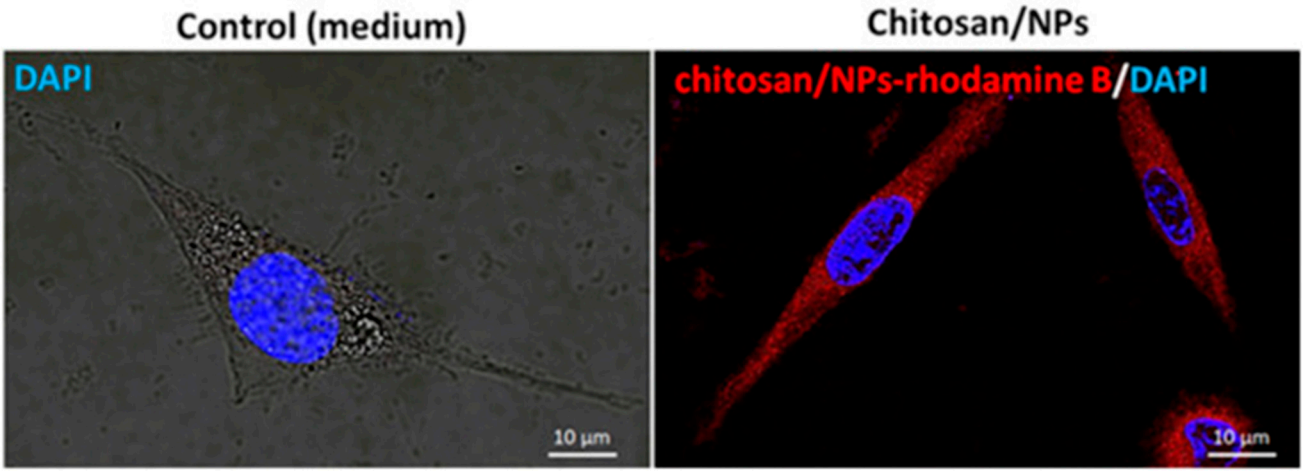

Figure 11. Images of L929 cells after incubation with untreated (control) or rhodamine B-loaded chitosan/NPs (with permission from the authors of [87]).

Even though sanitizers provide immediate hygiene benefits by disinfecting objects from microorganisms, they may damage electronic devices such as computers, refrigerators, etc. Thus, coating the objects that we use every day with antimicrobial films is considered to be quite an effective method of disinfection. Vitelaru et al. produced transparent antimicrobial films that were a few tens of nanometers thick and intended for use as screen protective films [2]. They placed these films on a self-adhesive polyurethane foil. SiO + TiO_2, SiO + Ag, and TiO + Ag compounds were used. TiO and SiO were used as a transparent matrix that was going to be embedded in silver nanoparticles; hence, the physical strength and the controlled development of the films were ensured. The accumulation time of SiO_2 and TiO_2 was approximately 1 nm/h, and the processing time was deemed to be 30 min. It was found that the transparency of SiO_2 film was the most effective one. However, most likely due to the insufficient amount of oxygen, it was observed that TiO_2 had the lowest amount of transparency, which led to stoichiometric compounds. Antimicrobial properties were evaluated by running a test using *Escherichia coli* strains. The maximum antimicrobial effect was observed in the Ag/SiO_2 combination.

Nazari et al. found out that, aside from TiO_2 and SiO_2, Ag nanoparticles could be combined with nepheline and used as an antimicrobial material in the production of ceramic tiles. In this study, $AgNO_3$ and PVP were mixed for two hours, and TiO_2, SiO_2, and nepheline were added to the prepared mixture.Then, the mixture of water and the solution was sprayed on ceramic tiles via a spray gun. The coated tiles and the traditional tiles were sintered. Iron dioxide created black spots on the surface, and this creates an undesirable effect on the thermal, chemical, and mechanical resistance of the tiles. After the antibacterial studies, it was found that the antibacterial activity (approximately 99.9%) of Ag/nepheline composite was higher than other composites. Consequently, the use of Ag/nepheline composites on ceramic surfaces was shown to be advantageous in terms of its structure and antibacterial properties [88].

Antibacterial agents used in the cleaning process of drinking water need to be nontoxic to humans, effective against a wide variety of pathogens, and cheap and easy to produce. One of the most productive and practical ways of placing these agents into a domestic environment involves coating the mixture apparatuses. Porel et al. transformed previously prepared thin Ag-PVA film polymers into a reusable water cleaner. For the production of silver nanoparticle-embedded polyvinyl alcohol (Ag-PVA) thin film, a simple previously developed technique was used including eco-friendly production protocol [89]. The sample of water indicated about 2500 CFU/mL; however, not a single colony was detected in the sample that was sunk into the film until the 20th use.

4. Antimicrobial Nanomaterial Coating Techniques

Biofilm formation and microbial adhesion are common, undesirable phenomena on the surface of living or non-living material in contact with microbial species. The deposition of antibiofilm coatings is one of the most popular approaches used to alleviate problems caused by biofilms on a variety of surfaces. Various coatings have been developed for the antimicrobial protection of surfaces. Of these, progress in the synthesis of graphene-based nanomaterials and composites with broad-spectrum antimicrobial activity and unique properties has created additional opportunities for the development of effective antimicrobial coatings. There are different methods for the deposition of graphene coatings, such as chemical vapor deposition (CVD), printed electronics, dipping, spinning, sputtering, and electrophoretic coating [90]. As an example, Janković et al. fabricated graphene-based silver/hydroxyapatite/graphene (Ag/HAP/Gr) composite coatings via electrophoretic deposition (EPD) on titanium. In this study, it was reported that the developed composite coatings showed antibacterial activity against *Staphylococcus aureus* and *Escherichia coli* and did not show cytotoxic activity against healthy peripheral blood mononuclear cells [91]. In their study, Gomes et al. evaluated whether the surface immobilization of graphene nanoplatelets (GNP) provides antimicrobial properties to Silicon rubber (SR). They used spray and dip coating to deposit a dispersion containing SR and GNP-M5 or its oxidized form (GNP-M5ox) on the silicon surface. As a result, the best antibacterial activity was observed on substrates obtained by immersion with low concentrations of GNP-M5ox, which showed lower bacterial adhesion and a higher percentage of death compared to silicone [92].

The coating of nanoparticles can be performed by using different techniques, such as electron beam evaporation, anodization, plasma spraying, dip coating, and sintering. Equipment coated via the use of these methods prevents bacterial colonization and reduces the integration of tissues and the adsorption of proteins. Anodizing is the process used to increase the coating of natural metal oxides on metal surfaces using electrolytic passivation. Similar to this process, recently, nanoparticles have been anodized on metal implants to prevent bacterial adhesion [93]. From these methods, a new implant surface modification approach was developed by Zhang et al via coating silver nanoparticles on the porous Ti surface via electron beam evaporation (EBE). The researchers adjusted the thickness of the silver nanoparticle coating on the porous Ti surface they prepared and optimized it by changing the duration of the EBE process. As a result, it was determined that composite porous Ti surfaces modified with silver nanoparticle coatings with EBE processes showed superior antibacterial properties [94]. In another study, Radtke et al. produced titania nanotube (TNT) coatings via the low-potential anodic oxidation of Ti6Al4V substrates in a potential range of 3–20 V. In this study, it was determined that the most suitable nanotubes for medical applications are TiO_2 nanotubes obtained using 5 V potential (TNT5). The TNT5 sample was found to have optimal biocompatibility properties, outperforming pure Ti6Al4V alloy and antibiofilm properties without enrichment with additional antimicrobial agents [95]. Roy et al. also used silver (Ag)/silver oxide (Ag_2O) doping in plasma-sprayed hydroxyapatite (HA) coating on titanium substrate in order to reduce bacterial adhesion. As a result of this study, it was determined that plasma-sprayed HA coatings doped with the optimum amount of Ag have excellent antimicrobial properties [96]. In a study by Page et al., titania and Ag-added titania coatings were prepared via the sol–gel dip coating method. As a result, the Ag-doped titania coatings were found to have significantly superior antimicrobial and photocatalytic properties compared to the titania coatings. In addition, Ag_2O–TiO_2 coatings have been found to be useful coatings for hard surfaces in hospital environments, as they are durable, reusable, and cleanable and have excellent antimicrobial properties [97]. In a study by Okazaki et al., titanium-doped hydroxyapatite (Ti-HAp) nanoparticles, which are dispersible and calcined at different [Ti/(Ca + Ti)] atomic ratios, were prepared using the anti-sintering method.

As a result, the researchers found that the number of *S. aureus* on the Ti-HAp-coated substrate was reduced by 43% compared to those on the normal HAp and original PET

coating [98]. Ghule et al. reported for the first time that ZnO nanoparticles can be coated on cellulose fibers of paper by using an ultrasonic approach. The researchers reported that this coating approach is compatible with green chemistry as it is inexpensive, simple, consumes minimal materials, and minimizes the use of solvent medium, unlike spray techniques and mechanical bench coaters. At the end of the study, it was found that the paper coated with ZnO nanoparticles exhibited antibacterial activity against *Escherichia coli* 11634 [99]. Konwar et al. studied biofilms with antimicrobial properties produced from a chitosan-iron oxide-coated graphene oxide nanocomposite hydrogel via the co-precipitation method. In this study, it was found that the chitosan-iron oxide-coated graphene oxide nanocomposite hydrogel films exhibited significant antimicrobial activities against Gram-positive and Gram-negative bacterial strains and that opportunistic dermatophytes had a non-cytotoxic nature [100].

5. Synthesis and Mechanism of Antimicrobial Nanomaterials

Nanomaterials can be synthesized through many different methods and interact with microbial cells through various mechanisms. Perhaps the most studied metal nanomaterials are Ag nanoparticles. With the development of nanotechnology, silver nanomaterials have been synthesized and have shown unique interactions with fungal and bacterial species. Various methods have been adopted for the synthesis of silver nanomaterials. Of these, conventional chemical and physical methods seem dangerous and very expensive. Among the various synthetic methods for silver nanomaterials, biological methods appear to be simple, fast, non-toxic, reliable, and green. A green chemistry approach for the synthesis of silver nanomaterials is very promising [101]. As an example, Nadagouda et al. performed the green synthesis of microwave-assisted silver nanostructures [102]. In another study, an environmentally friendly photo-irradiation-based process of antibacterial silver nanoparticles using *gui hua* leaves was reported by Ullah et al. [103]. In another study, Liu et al. synthesized silver nanoparticles in an environmentally friendly way using leaf extracts of *Nageia nagi* [104]. The production of integrated alginate fibers with AgNPs synthesized from natural Dolcetto grape leaves was also performed by Yu et al. [105]. Although the antimicrobial mechanism of silver nanomaterials has been extensively investigated, it still remains unclear. It has been found that silver ions interact with the peptidoglycan cell wall, causing structural changes, increased membrane permeability, and cell death, and silver nanomaterials can interact with exposed sulfhydryl groups in bacterial proteins, avoiding DNA replication [106]. In contrast to its antimicrobial effects, silver nanoparticles cause toxic effects in higher cell lines, such as in humans, oysters, zebrafish, and rats. Evidence in rodents indicates that silver nanoparticles can accumulate in the body, cross the blood–brain barrier, and damage various tissues. In a study of human cells, it was concluded that silver may be genotoxic [107]. In addition, metal nanomaterials are highly advantageous as active antibacterial agents because their surface area is extremely large for their size and they can easily penetrate the biofilm and penetrate the entire depth of the biofilm. The exposure of bacteria to nanoparticles causes the inhibition of encapsulating exopolysaccharide and proteins. Titanium dioxide nanoparticles can bind to hydroxyl groups in the polysaccharide matrix of synthetic biofilms, causing irreversible adsorption. The deposition of iron oxide and silica nanoparticles on biofilms can be achieved via electrostatic forces. Aside from these mechanisms, other nanomaterials also have the ability to kill bacteria by releasing charged antimicrobial agents. Core quenching is also a spike strategy used to disrupt biofilm formation [108]. Despite their antimicrobial effects, the toxicity of nanomaterials also causes limitations for these materials. For example, at the cellular level, the cytotoxicity of TiO_2 nanoparticles is uncertain, but both toxic and non-toxic results have been reported [109,110]. The green synthesis of nano materials is of great importance because of its lower toxicity, as well as its economic, repeatable, and low-energy process. For example, in a study conducted by Pushpamalini et al., TiO_2 nanoparticular green synthesis was performed using four different leaf extracts [111]. There are also two contradictory results for the toxicity of gold nanomaterials. Some groups argued that

gold nanomaterials were not toxic, while others showed the presence of toxicity in their research [112]. Geetha and his colleagues found that gold nanoparticles synthesized by using the flower essence of *C. Guianensis* exhibited anticancer activity [113].

Despite the numerous advantages of antimicrobial nanomaterials in the biomedical field, there are many challenges in the application of antimicrobial nanoparticle (AMNP)-coated medical implants. Protein contamination on NPs or implant surfaces can cause changes in antimicrobial properties, and sterilization processes can contribute to these property changes. The production of AMNPs and AMNP-coated implants depends on several factors, including the properties of the NPs and bulk materials, cost-effectiveness, antimicrobial efficacy, and process complexity. In addition, further investigation of the polymicrobial populations causing implant-associated infections is needed during the evaluation of AMNPs as surface coatings. Although there are several barriers to applying AMNPs as surface coatings, enhanced collaboration between clinicians, researchers, regulatory parties, and those in the industry could contribute to the development of nanoparticles for medical implants. However, the biological activity of nanometals can be modified to bring new properties for existing treatments. Multi-metal and metal oxide nanoparticles (metal-doped nanocomposites, alloys) are very promising in solving problems such as agglomeration or cytotoxicity in pure metal and metal oxide nanoparticles. Surface modifications can also be used to improve the properties (anti-caking and hydrophilicity) and biological activity of metal-based nanostructures. To reduce the toxicity of metal/metal oxide NPs, doping with other metal ions can also be used to improve the properties of nanostructures [114,115].

6. Discussion

Antibiotics have traditionally been used in medical practice to control microbial abundance and spread. Yet, the emergence of antibiotic resistance has led clinicians to be more cautious for the writing of medical prescriptions in order to decrease the overuse/abuse of antibiotics.Recently, there have been remarkable research developments regarding preventing bacterial contamination and biofilm formation by killing microbes or reducing their adhesion. These have been accomplished through the development of passive pathogen-repellent surfaces, biocidal coatings, surface-bound active antimicrobials, and micro-nanostructured surfaces. Antimicrobial surfaces, which can be created utilizing antimicrobial nanoparticles, appear to be a potential solution to prevent and reduce disease transmission. Nanoparticles and structures have demonstrated themselves to be efficient antimicrobials due to their high surface reactivity, small size, and diverse modes of action. The most common antimicrobial nanostructures in use today are made of silver, followed by antimicrobial peptides, chitosan, zinc, titanium, copper, and other materials. On the other hand, given that each virus has a different way of interacting with a surface, creating effective antiviral materials may require specialized customization for each virus type. While creating an antiviral substance, each of these factors should be considered individually and collectively. In order to promote interactions that can stop viral propagation, smart nanostructures on the same scale as and with a comparable geometry to viruses can be created. The current review aimed to summarize the state of research on antiviral surface coatings that may be used to stop the spread of pathogens during viral pandemics, as well as the difficulties and problems that still need to be resolved.

7. Conclusions

It is necessary to employ strategies that reduce microbial populations in hotspots where infections are prevalent because the unchecked, accelerated proliferation of potentially harmful bacteria can have detrimental effects. Surfaces become a major site of pathogen transmission because disease-causing microorganisms may persist on them for up to a few days and spread through them often. The areas of application where antimicrobial surfaces have been utilized most commonly include internal surfaces of the human body, such as catheters and implants, as well as external surfaces, such as marine surfaces, textiles,

paints, surfaces near to or used for food, and medical equipment. This review provides a comprehensive overview of nanomaterial-based antimicrobial coatings that can be used to stop the spread of contamination to surfaces.

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