



# ZnO-based antimicrobial coatings for biomedical applications

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## Abstract

Rapid transmission of infectious microorganisms such as viruses and bacteria through person-to-person contact has contributed significantly to global health issues. The high survivability of these microorganisms on the material surface enumerates their transmissibility to the susceptible patient. The antimicrobial coating has emerged as one of the most interesting technologies to prevent growth and subsequently kill disease-causing microorganisms. It offers an effective solution a non-invasive, low-cost, easy-in-use, side-effect-free, and environmentally friendly method to prevent nosocomial infection. Among antimicrobial coating, zinc oxide (ZnO) stands as one of the excellent materials owing to zero toxicity, high biocompatibility to human organs, good stability, high abundancy, affordability, and high photocatalytic performance to kill various infectious pathogens. Therefore, this review provides the latest research progress on advanced applications of ZnO nanostructure-based antibacterial coatings for medical devices, biomedical applications, and health care facilities. Finally, future challenges and clinical practices of ZnO-based antibacterial coating are addressed.

**Keywords** Zinc oxide · ZnO · Antimicrobial coatings · Nosocomial infection · Coating technology

## Abbreviations

ZnO	Zinc oxide	ROS	Reactive oxygen species
SARS-CoV-2	Severe acute respiratory syndrome coronavirus 2	DNA	Deoxyribonucleic acid
COVID-19	Coronavirus disease 2019	P(DMDAAC-AGE)	Polymer dimethyl diallyl ammonium chloride-allyl glycidyl ether
HAI	Hospital-acquired infection	HDTMS	Hexadecyltrimethoxysilane
Fig.	Figure	WCA	Water contact angle
UV	Ultraviolet	TEOS	Tetraethyl orthosilicate
NPs	Nanoparticles	SS	Stainless steel
VB	Valence band	NW	Nano-wall
CB	Conduction band	h	Hours
TM	Transition metal	PEEK	Polyether-ether-ketone
eV	Electron volt	NWs	Nanowires
RE	Rare earth	CMS	Carboxymethyl starch
DFT	Density functional theory	YAG	Yttrium aluminum garnet

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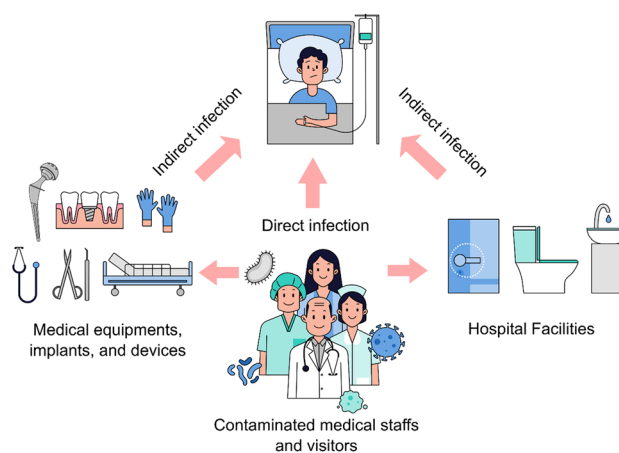
## Introduction

Surface coating plays an important role in determining the materials' functionality. Nowadays, surface coatings have been developed to possess even both decorative and functional properties [1]. While decorative properties enhance aesthetic appearance, functional properties provide a protective barrier against environmental hazards such as corrosive gas and chemicals, mechanical and thermal damages, or high-energy light exposures. In many industrial processes,

such as petrochemical and power generation plants, where many components and machinery come into a contact with a variety of gas and chemicals over a period of time, surface protective coatings are indispensable to protect critical parts from corrosion and extend their lifetime [2, 3]. The recent severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) pandemic, or well-known as COVID-19, has also brought out the importance of surface coatings as one of the safety measures [4]. The virus is transmitted via airborne respiratory droplets from coughing, sneezing, talking and even fomites [5]. A recent study showed that the coronavirus could survive for several days on fomite made of metals, polymers, or ceramics [6]. When a human is in contact with fomite without protective equipment, the virus may potentially be transferred to the human body through the nasal or mouth [5, 7]. Therefore, antiviral surface coating research will be of future interest in the prevention of the wide spreading of the viruses alongside vaccination and quarantine efforts [4, 6].

Not only viral infection but bacterial infection through the fomite of medical facilities have also been gaining vital concern across the globe [8, 9]. This is because modern healthcare uses various invasive devices and operations to treat patients and assist them to recover. Therefore, there is a considerable risk for the patients. This type of infection is known as hospital-acquired infections (HAI) or nosocomial infection [10]. As previously mentioned, nosocomial infection is caused by bacteria or viruses and typically incubating at the time of admission [11, 12]. All treated patients are susceptible to contracting an HAI and people with immature immune systems, such as infants and elderly have a higher risk of experiencing this infection [13]. According to Centers for Disease Control and Prevention (CDC), there are several types of HAI including central-line-associated bloodstream infection (CLABSI), catheter-associated urinary tract infections (CAUTI), ventilator-associated pneumonia (VAP), and surgical site infections (SSIs) [14]. In fact, more than 30% of HAI and 45% of CAUTI fatality cases induced by gram-negative bacteria which has ability to have a genetical code mechanism against antibiotic drug. The pathogenic bacteria originate from indoor area, surgical equipment, health staff clothing, etc. Moreover, the transmission media of pathogens include hospital staffs, hospital visitors, medical equipment with improper sterilization and inadequate high-level disinfection (HLD), and bad air circulation facilitate the spread of the bacteria-assisted nosocomial infection [15]. A schematic illustration of transmission routes of HAI is shown in Fig. 1.

A preventive and general measure of HAI is to perform routine cleaning and disinfection of fomites which probably limit the breed of microorganisms [13, 16]. However, the adopted sterilizations are not effective as some pathogen can survive on the surface of objects up to 5 months, such as *methicillin-resistant staphylococcus aureus* (MRSA),



**Fig. 1** Schematic illustration of transmission routes of hospital-acquired infections (HAI) or nosocomial infection

*vancomycin-resistant enterococcus* (VRE), *norovirus*, *Clostridium difficile*, *Acinetobacter species*, and *Escherichia coli* [10]. Furthermore, the overuse of disinfectants may produce multiple antibiotic-resistant bacterial strain. In relation to seek a non-invasive, low-cost, easy-in-use, side-effect-free, and environmentally friendly method to combat nosocomial infection, antibacterial coatings offer a bright promise [17, 18]. Its ability to directly kill and destruct bacteria and viruses may still be inferior to copper alloy-based materials which have been proven killing 99.9% virus and bacteria within couple of minutes [19]. Yet, copper-based materials are relatively expensive, visually inartistic, burdensome to coat them into substrate and thus they are not widely applied in medical facilities today [20]. Alternatively, ceramic-based antibacterial coatings have shown a different antibacterial mechanism, where the inactivation of bacteria and viruses can be activated with a light [21, 22]. In short, photon energy induces photocatalytic process to generate serial of non-selective microbial toxic active species (radicals, oxygen ions, peroxides) that have high toxicity to bacteria and viruses [23]. This adds to simplicity in sterilization of medical facilities [24]. Ceramic-based coating properties may meet the high provision of antibacterial coatings in medical devices due to their lightweight, inexpensive, toughness and hardness to mechanical damage, inertness to chemical and colorable [25].

Among the photoactive ceramic materials, zinc oxide (ZnO) nanostructures have a long historical study and development for their excellent antibacterial activity [26–29]. ZnO is an earth-abundant compound, and it exhibits great optical absorption near UV region, transparent conductivity, non-toxicity and biocompatibility to human organs, which make them particularly attractive for photodetectors, photocatalysts, cosmetics, pigments, and biomedical application [30–36]. With the advanced structural modifications, such as

doping, nanocomposites, etc., the antibacterial coating properties of ZnO can be further optimized [36–38]. There are number of published reviews on ZnO nanoparticles (NPs) for antibacterial agents, including their synthesis, characterizations, antibacterial mechanisms, and applications [21, 24, 39–43]. However, these reviews only highlight the antibacterial properties of ZnO in the nanoparticulate form, and a comprehensive discussion on surface-active antibacterial coating based on ZnO NPs has not been reported. Therefore, this review aims to show the potential applicability of ZnO nanostructure-based coatings in medical devices and health care facilities. With the rapid development of biomaterials science, it is urgent to stimulate practical relevance of antibacterial coatings for clinical adaptation in combating nosocomial infection. In this review, we also provide an insight into well-established coating technologies that may be suitable for manufacturing ZnO thin film. The critical parameters that significantly influence the quality of ZnO coating has been highlighted, clearly demonstrating that manufacturing methods are substrate dependent. Common failures in ZnO coating are also briefly discussed to increase awareness and improved processes from coating manufacturers. In author's view, to allow ease of understanding, this paper is divided into four main sections. First, an overview of properties of ZnO and historical development for the use of antibacterial coating is discussed, with a particular emphasis on performance comparison with other metal oxides. Second, brief photoinduced antibacterial inactivation mechanism is introduced. Third, antibacterial coating activity of ZnO on various substrate including metals and alloys, polymers and plastics, papers, textiles, and fiber as well as transparent glass and opaque ceramics. The section discusses key issues, required properties and corresponding performances of ZnO NPs which is for their practical relevance. Fourth, in terms of surface coating and manufacturing, various fabrication methods and parameters for surface coatings of NPs are compared. Fifth, opportunities and challenges for clinical application of ZnO nanostructure-based antibacterial coatings have been provided. Finally, we summarize the importance of ZnO antibacterial coatings applications in medical devices and health care facilities.

## ZnO as a photocatalyst material for bacterial inactivation

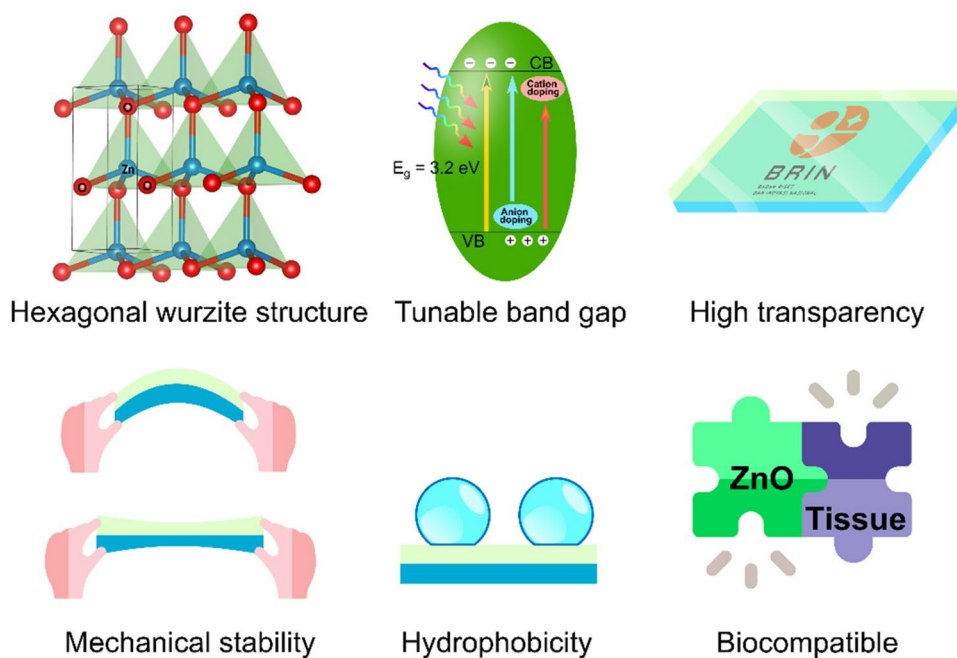
Semiconductors are well-known as the most potential photocatalysts due to their excellent physicochemical properties such as tailorable bandgap, high stability, and well-defined electronic configuration with occupied valence band (VB) and unoccupied conduction band (CB) that support the photocatalyst efficiency [44–47]. Given their great light absorption, charge transfer properties, and

adjustable morphological structure, photocatalytic activities for bacteria degradation can be expected. As a matter of fact, these materials are more durable than organic antimicrobial agents, can withstand adverse processing condition and chemically stable in aggressive environment (high temperature and pressure condition) [48, 49]. Photocatalytic disinfection has become an efficient way for inactivation of various pathogens such as bacteria, viruses, fungi, etc. to potentially inhibit nosocomial infection, because it utilizes the abundant and natural sunlight. In the last decades, transition metal (TM) semiconductors photocatalyst can be potential candidates as inactivation microbial species, and the followings are the most recognized and studied materials: TiO<sub>2</sub>, ZnO, WO<sub>3</sub>, MoO<sub>3</sub>, and CeO<sub>2</sub> [50–55]. However, some of the semiconductors corrode and degrade during photocatalytic reactions thus resulting the dissolution of semiconductor materials in the reaction system. For that reason, the highly stable excellent photocatalysts are most required.

As stated above, ZnO is classified as an n-type semiconductor and among the most investigated materials for antibacterial and biomedical applications due to the following advantages [34, 56]: (i) good chemical stability, (ii) tunable band structures, (iii) high optical transparency, (iv) excellent mechanical stability, (v) surface hydrophobicity, (vi) low production cost, (vii) low toxicity and high biocompatibility, and (viii) possessing excellent photocatalytic antibacterial inactivation, as depicted in Fig. 2. Herein, we start with ZnO crystal structures. In common, the crystal shapes of ZnO are cubic rock-salt, hexagonal wurtzite, and cubic zinc-blende [34]. The hexagonal zinc wurtzite is the most thermodynamically stable under ambient conditions. ZnO with wurtzite structure is consisted of C<sub>3v</sub> point group with each O<sup>2-</sup> ion which is bonded by four Zn<sup>2+</sup> ions at the corners of a tetrahedron, and likewise each Zn<sup>2+</sup> ion is coordinated by four O<sup>2-</sup> ions in a tetrahedral arrangement. The wurtzite structure has lattice parameters of  $a = 3.2495 \text{ \AA}$  and  $c = 5.2062 \text{ \AA}$  [34]. It is well-known that the sp<sup>3</sup> hybridized bonds of almost equally ionic and covalent character are the main chemical bonding in ZnO crystal [34]. The unstable polar symmetry situated around [0001] plane terminated by Zn-atom, and another arranged along [0001] plane terminated by O-atom on its surfaces. These two directions are additionally affecting their properties. Besides that, there are some steady and non-polar faces, for example, [0110], [0110], [1100], [1100], [1010], [1010], that favor to obtain various morphologies of ZnO nanocrystal with different dimensionality [57].

Wurtzite structure ZnO is n-type semiconductor with a direct bandgap of 3.37 electron volt (eV), which gives high transparency in the visible light region and is suitable for direct UV utilization optoelectronic devices [58]. The n-type conductivity is originated from the intrinsic defects, such as Zn interstitials (Zn<sub>i</sub>) or O vacancies (O<sub>v</sub>) [33, 59], although more recent works proposed that the Zn vacancies (Zn<sub>v</sub>)

**Fig. 2** ZnO crystal structures and its electronic, optical, mechanical, surface, and biocompatibility properties



are likely responsible for the n-type conductivity, because they are acting as deep acceptors to compensate the n-type charges [60, 61]. Besides, possessing large formation energy makes  $Zn_i$  shallow donors unstable [62]. The relatively wide bandgap limits the visible-light-induced photoactivation antibacterial properties. This can be overcome by the incorporation of various atoms the ZnO lattice or well-known as doping. It has been proven that the introduction of dopant has induced to the ferromagnetic and electromechanical behavior of the ZnO. For a clear and comprehensive discussion, we divide the elemental dopants into cations and anions. Cations are the most studied elements in the field of doped-ZnO. Many cations are well-suited to the ZnO crystal and incorporated by replacing the Zn sites or inserting into interstitials lattice. For example, rare-earth (RE) elements (Tb, La, Gd, etc.) allow the ZnO to prolong the recombination of photogenerated electron/hole pair due to the 4f configuration, maximizing its quantum efficiency. Nevertheless, most of RE elements have larger ionic radii than the Zn ion. This can be a disadvantage on one side, because the large difference in ionic radii causes low dopant incorporation and may destroy the pristine morphology of ZnO. In another side, the large ionic radius may induce a stronger distortion of the crystal cell and the rise of a stronger spontaneous polarization to wake up the ZnO piezoelectricity. So far, only Nd-doped ZnO showed the enhancement of the antibacterial activity [63].

Another cations dopant from the TM elements have also been investigated. There are many studies demonstrating the successful incorporation of TM elements, such as Fe, Ni, Cr, Mn, Cu, Co, Ti, and so on into the crystal structure

of ZnO. The biggest advantage of doping with the TM is their ionic radii similarities [64]. This can ensure the high solubility and homogeneity of the TM elements in the ZnO crystal lattice without altering the parent particle size and morphology [65]. Some of TM will induce the magnetic and piezoelectric properties of the ZnO such as by Fe, Cr, or Mn due to the spin coupling and the asymmetric wurtzite structure formation, respectively [66]. The use of Fe, Cu, Ni, Mg, and Ta is effective to increase the bacterial growth inhibition and antibacterial properties due to the large number of reactive oxygen species (ROS), cytotoxic zinc, and copper ions generation [22, 67, 68]. The antibacterial mechanisms are discussed in the next section. Apart from RE and TM, the noble metals element such as Ag and Au has also been successfully incorporated into the ZnO, giving higher antibacterial properties due to the same effect which TM gives [69]. Nevertheless, due to excessive cost of the noble metals, their use is not preferable in making cost-effective ZnO antibacterial coating. The actual choice of cations doping is very broad ranging from the alkali metals (groups I and II) and group III (Al, Ga, In) to d-orbital transition metals (W, Mo, Ru, Ta) and dichalcogenides (Se, Te) [70, 71]. Because of this fact, wide variety of cations-doped ZnO can potentially be produced with the enhanced properties depend on type the dopant and doping concentration. In most cases, introduction of foreign cations into ZnO wurtzite lattice reduces the bandgap energy, because the dopant level is below the CB, where photogenerated charge carriers are trapped. This approach is effective in enhancing microbial growth inhibition properties of ZnO.

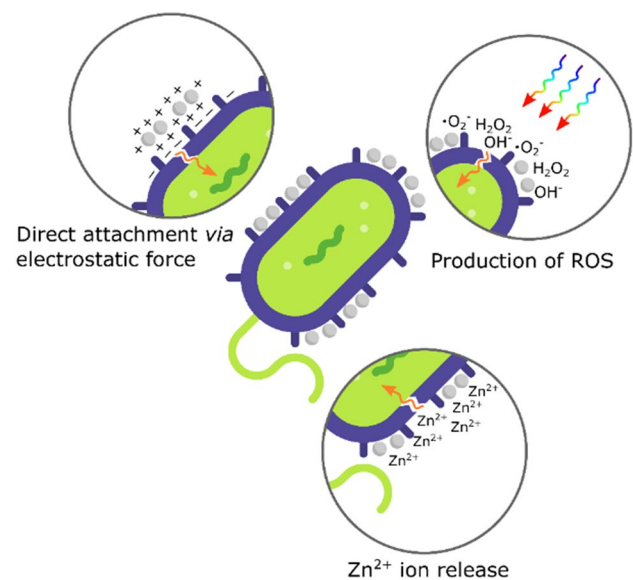
Likewise, the antibacterial performances of ZnO can also be improved by introducing anions dopant (N, S, C, F, and Cl) into ZnO lattice. Anion dopants will substitute the lattice oxygen atoms, because of their atomic radius similarity and lower electronegativity compared to oxygen [72]. Specifically, N atom offers a similar  $2p$  energy state, high solubility in ZnO, and induces oxygen vacancies. The hybridization of N  $2p$  and O  $2p$  orbitals repulses the Zn  $3d$  orbital, narrowing the bandgap of ZnO from 3.4 eV to 3.1 eV [73]. Besides the p–d repulsion, the formation of defect states account for the high antibacterial activity of N-doped ZnO materials, as the defect formation increases the amount of ROS [74]. A decrease in bandgap was also observed in C-doped ZnO and S-doped ZnO. According to the density functional theory (DFT) calculation, a new band formation positioned at the top of VB level due to the orbital hybridization has induced the energy band shifting downwards [75, 76]. In addition, the formation of oxygen vacancies provides a trap to retard the  $e^-/h^+$  recombination. However, the larger ionic radii of  $S^{2-}$  and  $C^{4-}$  may deteriorate the desired morphology and particle size of ZnO because of the lattice expansion. Thus, a careful consideration is required when incorporating S and C dopants. We also refer readers interested in the more detailed improvement strategies of the photocatalytic and antibacterial activities of ZnO to the available literatures [39].

### Mechanisms of antibacterial activity regarding ZnO-based coatings

For many years, researchers have been investigating working mechanism of oxides-based photocatalytic antibacterial activity, where ZnO is used as a general model due to its excellent antibacterial performances. Although the primary antibacterial mechanism is still unknown, it is true that the nature of antibacterial activity of ZnO has been fundamentally corresponded to their photocatalytic properties and these following mechanisms are well-acknowledged to explain its antibacterial performances: (i) active release of  $Zn^{2+}$  ions from ZnO nanocrystal, (ii) production of reactive oxygen species, and (iii) direct attachment between ZnO NPs and bacteria cell via electrostatic interaction-destroyed bacterial cell integrity followed by the malfunction of cell membrane by infiltration of ZnO NPs, as illustrated in Fig. 3. Herein, each mechanism is explained and compared to enlighten the effective and efficient design of antibacterial coating based on ZnO nanoparticles, which can extend the medical applications for ZnO.

#### Formation of $Zn^{2+}$ ions

Previous comprehensive works have led to a well-balanced conclusion that antibacterial activities and toxicity of ZnO



**Fig. 3** Various antibacterial mechanisms of ZnO toward Gram-positive and Gram-negative bacteria

are strongly associated with the particle dissolution into  $Zn^{2+}$  ion. In this aspect, the  $Zn^{2+}$  ions play critical role in the formation of ionic signals between in catalyst and protein arrangements between different cells and intercellular cytoplasmic organoids [77]. It is also believed that  $Zn^{2+}$  ions are of importance in inhibiting active transport, metabolism of amino acid, and enzymatic disruption [78]. Huang and co-workers have found interesting phenomenon from their study on bacterial inactivity (*Streptococcus agalactiae* and *Staphylococcus aureus*) by ZnO nanoparticles, in which the dissolved ions were very destructive to bacterial cell and membrane—that is increasing possibility of ZnO NPs penetrating into cell [79]. In fact, metal ion homeostasis is maintained through highly regulated processes of uptake, storage and secretion, and significant for bacterial life, since they are engaged with various supersession purposes, for example, dehydrogenase, cofactor and catalyst agent, as well as balancing out specialist for chemicals and deoxyribonucleic acid (DNA) binding proteins [80]. Nonetheless, exaggerated metals or metals ions are harmful to bacteria cells. Specifically,  $Zn^{2+}$  ions released from ZnO NPs provide more toxicity to bacteria compared to ions formation from  $TiO_2$ , CuO, NiO, etc. [81]. Furthermore, some bacteria have showed some mechanism to maintain in draught and outflow of metal ions such as  $Zn^{2+}$  to regulate stable intracellular ion concentration [82]. ZnO NPs with positive zeta potential can annihilate the cytomembrane of gram-negative *E. coli*. Reacting ZnO powder with bacteria will likely induce the slow release of  $Zn^{2+}$ . Regarding to ZnO oxidation capacity,  $Zn^{2+}$  not only responses to organic functional group (thioglycoside, carboxyl, hydroxyl), but also merge to the

bacteria cell and membrane protein. It goes into the bacteria cell and harms its electron move framework to reject the enzyme and protein gene expression function, which are produced in the targeted antibacterial [83]. Thus, irreversible damage occurs in all internal bacteria organs by active absorption of  $Zn^{2+}$  ions.

Still, there are controversies exist regarding to which extent the  $Zn^{2+}$  contributes to the underlying mechanism of antibacterial properties of ZnO nanoparticles. A comprehensive and systematic research demonstrated by Bellanger et al. have come into conclusion that no quantitative relationship between the released ions of  $Zn^{2+}$  and their antibacterial activities toward *E. coli* MG1655 and *Cupriavidus metallidurans* CH34 [84]. Despite some studies did not show a good agreement with this mechanism, they do not necessarily eliminate the role of dissolved  $Zn^{2+}$  ions toxicity. It may originate to the nature of bacteria itself when interacting with ZnO NPs or  $Zn^{2+}$ . Some bacteria may exhibit more sensitivity than other bacteria. Moreover, alteration research media and components in which the antibacterial testing is conducted, cannot generally be avoided. In this regards, comprehensive works need to be performed as soon as possible.

### Production of reactive oxygen species

Another proposed mechanism which is responsible for antibacterial properties of ZnO nanoparticle is generation of ROS which has been demonstrated in several works [26]. Not only in ZnO nanoparticles, but the photogenerated ROS can also be produced in most of engineered metal-oxide nanoparticles [85]. Therefore, it is thought to be the primary mechanism of excellent antibacterial activity of ZnO NPs against both Gram-positive bacteria and Gram-negative bacteria. In principle, by the help of light illumination with photo energy ( $h\nu$ ) larger than the bandgap of ZnO, the electrons ( $\bar{e}$ ) and holes ( $h^+$ ) in the VB of ZnO NPs are separated, where ( $\bar{e}$ ) promote to CB and holes ( $h^+$ ) moves downward of VB (Step 1) [86]. Due to high oxidizing property, the photogenerated holes deteriorate the atmospheric  $H_2O$  molecules into hydroxyl radicals ( $\bullet OH$ ) and protonated hydrogen ( $H^+$ ) in the ZnO suspension (Step 2). Meanwhile, the photoinduced ( $\bar{e}$ ) react with molecular oxygen, through reductive process, to form superoxide anion radicals ( $\bullet O_2^-$ ), which then react with ( $H^+$ ) to generate hydroperoxyl radicals ( $HO_2\bullet$ ). Singlet oxygen ( $^1O_2$ ) is also a strong oxidative agent and is indirectly produced from aqueous reaction of ( $\bullet O_2^-$ ). In the final step, hydrogen peroxide ( $H_2O_2$ ) molecules are formed by the recombination of ( $HO_2\bullet$ ), ( $H^+$ ) and ( $\bar{e}$ ). Each product is believed to have biochemical induction processes with bacteria.  $\bullet OH$  radicals are the most reactive and nonselective oxidant, where they can destroy most of the biomolecules (carbohydrates, nucleic acids, lipids,

amino acids, etc.).  $^1O_2$  is responsible for phototoxicity and destroying the surface of microorganism membrane and degrading the integrity of the bacteria cell through the oxidative stress process [87]. Many studies have proved that the three ROS ( $\bullet OH$ ,  $^1O_2$ , and  $\bullet O_2^-$ ) largely contribute to the membrane lipid peroxidation via oxidative stress process in many biological systems, although the latter ROS is not equally strong. In addition,  $H_2O_2$  is the important part to affect antibacterial activity of ZnO nanomaterial [88]. The formation process of  $H_2O_2$  on the surface of ZnO NPs is related to oxygen species.  $H_2O_2$  is able to penetrate into the cells of weak electrostatic interconnection, causing the dysfunction of DNA, cellular proteins and death of molecules [89]. The surface grain and size of ZnO NPs clearly regulate the amount of  $H_2O_2$ , where smaller NPs generally exhibit high content of  $H_2O_2$  and oxygen species on the surface which logically have higher antibacterial performance [90]. However, it is still under debate whether generation of ROS can only be triggered by photon energy. Some studies have provided compelling evidence that ZnO exhibit equivalent antibacterial properties in the absence of light [91]. There must be co-existed mechanism that responsible for toxicity of ZnO against bacteria. The main possibility is that pre-existed oxygen defect in n-type ZnO lead to electron-rich crystal to produce ROS with similar mechanism in step 3 [92]:



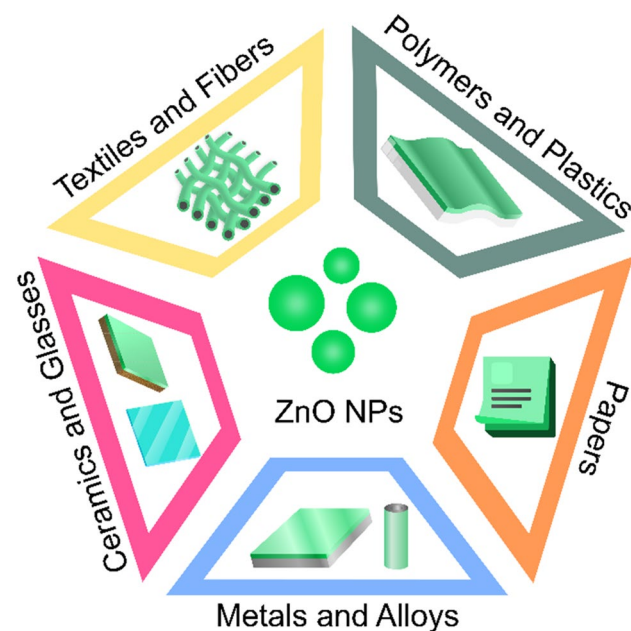
### Direct attachment of ZnO NPs and bacteria cell via electrostatic interaction

Another potential mechanism to explain antibacterial activity of ZnO is that direct interaction between ZnO NPs and exterior of bacteria via electrostatic force. The total effective surface charge is indicated by the zeta potential value. While many bacteria possess a net negative charge as a result from carbonyl group, ZnO nanoparticle maintains its positively charged in the suspension contains of water (pH is about 7) [93]. Such opposite charges can induce a strong electrostatic force interaction, acting as physical bonding for ZnO to surround the bacteria cell. Consequently, the cell is damaged and the entire bacteria

is dead through apoptosis [94]. Using bio-transmission electron microscope (TEM) observation, one can observe that the attachment of ZnO on the outer cell of bacteria has induced pits formation, in which the released lipopolysaccharides inhibit bacterial growth. ZnO can slowly penetrate through the pits, particularly at the cytoplasmic site and periplasm space, that could be able to obstruct cellular metabolism and membrane disorder [78, 94].

## ZnO-based antibacterial coatings

ZnO antibacterial coating has been used in many applications such as in food packaging, textile, biomedical application, etc. As we know, hospital facilities contain many bacteria, where they are more resistant to antibiotics and external forces and can withstand host immune responses. The antibacterial coating prevents the bacterial colonization of material surface and acts as permeation barrier coating, besides those thin films can impart desired surface functions without affecting bulk mechanical properties. To this end, inhibiting various microorganisms in healthcare facilities, ZnO should be able to suitable coated and integrated onto different substrate, as shown in Fig. 4. In the following section, we summarize the recent progress of ZnO antibacterial coating properties on different surface which classified as (i) textile and fibers (ii) metals and alloys, (iii) polymer and plastics, (iv) transparent glass and ceramics, and (v) papers.



**Fig. 4** ZnO antibacterial nanocoating on different substrates. Green color represents antibacterial coating based on ZnO NPs

## ZnO NPs-based antibacterial coatings on textiles and fibers

Healthcare facilities commonly use textile or cotton fabrics in their appliances, e.g., patient dresses, bed cover sheet, blanket, table cover, and so on. The spread of bacteria and viruses on the surface of the textile and cotton fabrics become one of the problems which can initiate the HAI. ZnO nanostructure has been widely approved as a chemical agent-coated textile because of its unique physical and chemical properties, environmental friendliness, biocompatibility, low toxicity, and low-cost production; thus, many personal-care products incorporate ZnO as one of the active components. Furthermore, as natural fibers can be vulnerable to surrounding damages induced by microorganisms and UV light irradiation, the increased surface properties and the durability of natural fibers can be expected when they are protected by ZnO NPs coatings.

Petkova et al. have successfully synthesized ZnO thin films on medical textiles using a simultaneous sonochemical-enzymatic coating method [95]. The method provided better ZnO NPs adhesion to cotton fabrics, which consequently improve the coating stability during the sample testing against *E. coli* and *S. aureus*. ZnO NPs-coated fabrics has effectively inhibited the growth of those bacteria by 67% and 100%, respectively. The excellent antibacterial activity was remained effective after multiple washing cycles with a non-ionic detergent in hospital laundry condition, although only 33% of ZnO NPs remain intact on the cotton fabrics. The sono-enzymatic has offered an alternative approach to overcome the intensive chemical treatments used in modern industry of textile coating, yet the critical issue of ZnO NPs adhesion stability should be addressed.

To stabilize the ZnO NPs adhesion on the cotton fabrics and prevent the particle agglomeration, the polysiloxane (or known as silicon) network can be combined with ZnO<sub>2</sub> NPs to form ZnO/SiO<sub>2</sub> hybrid nanocomposite-coated cotton fabric [96]. To obtain the well-coated cotton fabrics, the cottons should be immersed in the sol precursors containing tetraethyl orthosilicate (TEOS) and zinc acetate mixtures, otherwise the agglomeration was still be observed. SiO<sub>2</sub> coating can act as a stabilizing agent and enhance the stability of ZnO nanoparticles. As expected, the inhibition zone of ZnO/SiO<sub>2</sub> against *S. aureus* and *E. coli* was wider with a more uniform and stable coatings. Although the effect of SiO<sub>2</sub> network on enhanced antibacterial activity mechanisms is poorly investigated, the generation of ROS and Zn<sup>2+</sup> release was likely to be dominant mechanisms in the present case.

Controlling the concentration and particles size of ZnO NPs on the cotton fibers are prominent in influencing their antibacterial coating activity, i.e., the higher concentration and smaller particle size of ZnO could increase antimicrobial activity, which has been well-approved by scientist

community. The hydrophobicity nature of ZnO NPs-coated cotton fabrics should be taken into primary consideration, as ZnO should repel the bacterial-contaminated water moisture or droplets by itself. The hydrophobic behavior of ZnO-NPs is commonly influenced by the composition of coating materials, the synthesis method and morphology of ZnO. The composition design can be performed by mixing ZnO NPs with polymers to form organic–inorganic nanocomposites. The P(DMDAAC-AGE)/Ag/ZnO composite was synthesized by free radical polymerization, where the epoxy groups were utilized to form covalent bonds with the hydroxyl groups of cotton fabrics [97]. The organic–inorganic nanocoating can protect the fabrics from the growth of *E. coli* and *S. aureus* up to 99.7% and 99%, respectively, in which the antibacterial properties can be retained after several laundry washing. However, it needs special consideration with the possibility of the polymer leaching which can be a potential water contaminant. Within the context of synthesis methods, Rilda et al. have synthesized the in-situ growth of hexadecyltrimethoxysilane (HDTMS)-modified ZnO-SiO<sub>2</sub> coating using a chitosan template-assisted hydrothermal method at 95 °C. The chitosan has provided the platform for the ZnO-SiO<sub>2</sub> growth with a highly roughened surfaces, which can synergize with HDTMS to bring up the super hydrophobicity properties. It was further revealed that the water contact angle (WCA) depended on the number of HDTMS layers, where the WCA of 150° was reached with only two layers of HDMTS coating. The morphology of ZnO NPs will have a significant effect to the hydrophobic properties of the cotton fabric, where the rod structures exhibited higher WCA than flake and flower-like ZnO-NPs [98]. Regardless of the morphology, the ZnO-NPs have shown a high light absorption capability and excellent inhibition to many Gram-positive and Gram-negative bacteria. Furthermore, a large-scale process to coat the hydrophobic and antibacterial bed sheet with ZnO has been proposed using a rolling-dipping machine [99].

Despite all the interesting achievements related to the utilization ZnO NPs as a coating material in textile, there are still some challenges. For instance, mechanical degradation of ZnO coating on cotton fabrics has been revealed. The woven fabric has a degraded tensile and tearing strength both in warp and weft directions due to the ZnO deposition [100]. The tensile strength in the warp direction of ZnO-coated cotton fabric has been reduced to 94.5% [101]. It has been proposed that the nanoparticle properties, treatment, and fiber modification process among the principal factors influencing to mechanical properties of the coated fibers [102]. Although deposition of ZnO on fabric enhanced the antibacterial activity against various microorganisms, the decreased mechanical properties (strength, strain, and abrasion resistance) were also observed. Also, some studies showed that antibacterial properties can endure multiple

washing cycles; however, in reality, the wash durability is still a huge issue [103]. In this context, the use of some chemicals to stabilize ZnO coating may alleviate this issue. To note, the chemicals should provide smoothness and surface protection while maintaining the excellent antibacterial properties of ZnO. The stable ZnO coating on fabric is essential as it will increase the long-term serviceability of the modified textile.

### ZnO NPs-based antibacterial coatings on metals and alloys

The metals and alloys are essential parts of healthcare facilities and biomedical equipment: stents in blood vessel and heart valves, orthopedic implants, cardiovascular devices, and so on [104, 105]. Metals and alloys for biomedical application should have some basic requirements such as biocompatibility, high corrosion resistance, toughness, elasticity, low wear, and sufficient mechanical strength [106]. Titanium alloys, magnesium alloys, and stainless steel (SS) 316L are the most common metals being used in biomedical materials because of their distinguished properties [107]. Implant materials have critical problems related to bacterial infection which can propagating to extended hospitalization, failure of implant, complexity of correcting procedure, and chronic diseases. The metals and alloys with antibacterial coating features hold a promise to prevent several infections and avoid biofilm formation on the implant surface. ZnO NPs should possess biocompatibility, improved corrosion resistance and increase antibacterial performance against various living microorganism.

Stainless steel is a fundamental material in door handles, cutlery, operating tables, furniture, water pipes, faucets, and containers in health care facilities which are often being growing and spreading media for bacteria and viruses. The application of alcohol or alkaline solution to sanitize the surface of SS is not long-lasting to reduce the bacterial growth and kill the entire viruses. Thin-film antibacterial coating becomes reassuring solution to reduce the bacterial growth, but the challenge of toxicity of ZnO NPs coating become needs a careful evaluation. Shim et al. investigated the toxicity of ZnO nano-wall (NW) film on the skin using elution tests [108]. The short time ZnO exposure to the skin is a relatively safe and can be considered as non-irritant based on the primary irritation index (P.I.I). However, contact between ZnO and the skin for 24 h (h) and 72 h can cause skin irritation evidenced by inflammation, red spots, and a skin rash [108]. Based on these results, it is recommended to shorten the contact time between the skin and the ZnO NW-coated SS to prevent skin irritation and inflammation. The toxicity of ZnO NW film on drinking water was also investigated, since the ZnO coating may leach from metal substrates to water resources. The results showed the Zn



ion concentrations were  $20.9 \pm 2.4$  and  $27.2 \pm 3.0 \mu\text{g L}^{-1}$  at  $100^\circ\text{C}$  after 24 h in the elution test. The value of Zn ions on the drinking water is still in the safe value limit of WHO guidelines (under  $3000 \mu\text{g L}^{-1}$ ). Although the number of Zn ions in drinking water is still safe, but the research on the elimination of Zn ions is still being developed. Meanwhile, the SS with ZnO NW coatings has achieved 99.5% inhibitions for the growth of multiple bacteria, namely *E. coli*, *S. aureus* and *Penicillium funiculosum*.

The other factor which should be prioritized is the hydrophobicity of ZnO nanostructure thin film on the surface of metal, similar to substrates made of fabrics. The hydrophobicity feature is needed for the stability of coating against liquid and moisture. The hydrophobic properties have a positive repercussion on antibacterial properties of ZnO nanostructure coating on metals against *Shewanella putrefaciens* [109]. The ZnO coating induced the bacterial biofilm formation when the bacteria adhered to the surface in the first 12 h. The adherence of bacterial biofilm will increase as the increased the exposure time from 12–24 h and degenerated after 24 h. The hydrophobic phenomenon of ZnO nanostructure on metal surface could be an advantage in many applications requesting the bacterial biofilm prevention, such as food packaging, food processing equipment and other antibacterial functional material fields. To provide versatile solution of the insufficiently inhibition properties of ZnO NPs for some bacteria, the combination with other antibacterial active materials to form nanocomposites has been proposed. However, the use of metal as a combinatory material should be avoided to restrain the bacterial resistance. Introducing carbon (C), sodium alginate (Alg), polyvinyl alcohol (PVA), and bioactive glass (BG) to ZnO NPs coating were effective to inhibit the growth of *S. aureus* and *Salmonella enterica* although they showed poor resistance against *Pseudomonas aeruginosa* [110]. Besides, the addition of bioactive glass to the nZnO/Alg-PVA sample could increase bacterial growth after 1 h of incubation. This phenomenon caused by the low concentration of BG and ZnO and thus the appropriate variable should be considered to improve the properties of combined materials.

Orthopedic biomaterials are meant to be implanted inside the human body and they should be designed according to the tight regulation for the successful surgery process. Without a proper cleaning and sterilization, implant materials have some risks of infections which may lead to implant failure even chronic disease. The hygiene of the operation room is not the only determining factor in avoiding the infection in implant materials. The content of bacteria in surgery appliances and the implant material itself plays major roles [111]. The recommendation for these problems is using antibacterial surface coating which prevent bacterial adhesion on the implants. ZnO NPs can be coated on the implant as antibacterial protection and anti-corrosion

protection. For example, the PEO/ZnO NPs-coated AZ31B magnesium alloys implant exhibited reduction activity to *S. aureus* and *E. coli* colonies at pH of 13.2 [112]. The antibacterial performance can further be enhanced by illuminating the UV-irradiation to excite more electrons-holes pairs and generate radical species. With UV irradiation, the bacterial inhibition activities were  $94.50 \pm 1.25\%$  against *S. aureus* and  $98.95 \pm 0.71\%$  against *E. coli*, where the sample without UV irradiation was only  $82.47 \pm 1.41\%$  against *S. aureus* and  $67.70 \pm 1.32\%$  against *E. coli*. Therefore, the surface cleaning of PEO/ZnO-coated metal implants from microorganism can be performed with the aid of UV light irradiation. Hybridization of ZnO with chitosan has recently shown a promise in improving the mechanical and bioactive performance of titanium orthopedic implants against *E. coli* [113]. In a word, nanocomposites based on ZnO NPs coating, especially with the polymers, become great interest for biodegradable implants, dental implants, orthopedic implants, and other biomedical applications. Also, the using of biocompatible materials with low toxicity and high biocompatibility become very profitable, because they are not only well-suited with implant application but also holding a great promise for wound healing application.

### ZnO NPs-based antibacterial coatings on polymers and plastics

Polymer and plastics are broadly used in many applications such as in household needs, food packaging [114], and medical devices [115]. Because of the high potentiality of polymers, the development of polymeric materials with antimicrobial activity is continuously increasing in the recent years [116]. Polymers and plastics possess a lightweight, low cost, wear-resistance and high flexibility to some extent, and they are everywhere in medical facilities. To illustrate, face shield, syringes, tubing, prosthetics, and polymer bed sheet cover are made from polymers. The most common polymers include polyurethane, polyethylene, and polytetrafluoroethylene (PTFE), along with high-performance polymers like polyether-ether-ketone (PEEK). The using of ZnO NPs as antibacterial coating on that medical equipment could reduce HAI. However, we need to have ZnO NPs with high hydrophobicity and flexibility when they are applied as coatings on polymers and plastics. Herein, we highlight recent development of ZnO coating on these substrates.

As the one of the most contaminated equipment in healthcare facilities, bed sheet is usually covered by antibacterial bed sheet to reduce the spread of infections. Kurniawan et al. found out the importance of ZnO coating cycles to the hydrophobicity of polyvinyl chloride (PVC) bedsheets [99]. The hydrophobicity properties are critical in decreasing liquid materials penetration inside the bed sheet, resulting in the reduced bacteria transmission from

the patient's blood or other physical secretion, which is the largest source of bacterial transmission in healthcare facilities. The wettability testing was performed to ZnO NPs to prove the effectiveness of different coating cycles to hydrophobicity. The results showed that the higher cycles induced the higher hydrophobicity properties. Besides that, ZnO NPs-coated bed sheets showed better hydrophobicity than uncoated bed sheets due to the hydroxyl group of ZnO, which usually possesses the hydrophilic properties, i.e., water repellent. The value of contact angle in the uncoated bed sheet, 10 cycles, and 15 cycles of ZnO coating showed values, respectively, of  $87^\circ \pm 1.29$ ,  $110^\circ \pm 0.45$ , and  $117^\circ \pm 0.76$ . ZnO-coated bed sheet effectively inhibited the growth of several bacteria such as *S. aureus*, *S. epidermidis*, *E. coli*, and *P. aeruginosa* better than the uncoated bed sheets. The coating cycles are suitable for hydrophobicity and antibacterial properties, but the roughness and blistering of ZnO NPs coating should be considered for flexibility and better surface appearance. The washable and reusable features of ZnO-coated polymers is also a must for improving economic efficiency [117].

The flexibility of ZnO NPs coating is also an essential factor in the application of bed sheet cover and medical gloves. Li et al. have successfully fabricated the highly elastic ZnO NPs-reinforced polyurethane coating [118]. The advantage of polymer elasticity also affects the use of ZnO NPs coating in broader applications, such as polyethylene for food packaging. Almost biodegradable polymers such as starch, gelatin, and cellulose are lack antibacterial activity, so the use of ZnO NPs into starch-coated polyethylene could improve their antibacterial activity. Tankhiwale et al. revealed the increasing antibacterial activity of starch coated polyethylene (SCP) film due to the addition of ZnO NPs [119]. We can extend the application of the fabricated polymers for medical purpose. The approach is eco-friendlier and economically cheaper as compared to other metal NPs antibacterial agents, such as silver and gold.

Besides the advantages of bare ZnO NPs, ZnO-based nanocomposites have also become recent of interest in the biomedical and pharmaceutical industries. The combination with other materials could increase antibacterial activity, hydrophobicity properties, tensile strength, and self-cleaning properties. Wang et al. revealed that caprolactam-casein ZnO (CCZ) nanocomposite latexes showed higher flexibility, increased antibacterial activity, and hydrophobic properties of the casein film [27]. Adding ZnO NPs could increase their toughness due to their properties as mechanical reinforcement to the combined polymer. However, adding more than 2% of ZnO NPs could form agglomeration and aggregation, which finally decreases the mechanical properties of the nanocomposite. Based on the many investigations, the proper amount of incorporated ZnO NPs into polymer

matrix should be considered to prevent agglomeration and aggregation, and this will be the major task for future research.

### ZnO NPs-based antibacterial coatings on transparent glass and opaque ceramics

In common health care facilities, including medical devices, various glasses and opaque ceramics can be found in windows, computer and device screens, walls, surgical implants, prosthetics, orthodontics, and many medical tools, respectively. Besides retaining their excellent antibacterial performance from those bulk powders, essential requirements arise when ZnO NPs coating is applied to those substrates. While ZnO NPs should possess high transparency for antibacterial coating on transparent glass, it is less required in ceramics substrate unless otherwise specified (dental implant as one representative example). Instead, interface compatibility with the substrate and surface durability is more critical. Herein, we discuss recent development on antibacterial coatings of ZnO NPs onto transparent glass and opaque ceramics.

As one of the building materials that allow the light into the room and as a protecting barrier from outside weather, glass with hygiene properties is most desirable in hospitals and health care centers to not be contaminated by bacteria and viruses [120] monitors, screens, and touch panels of medical equipment demand highly comparable properties [121]. ZnO holds such promising properties, acting as transparent conducting oxide (TCO) as well as antibacterial and antimicrobial coating. Fabricated by various methods, ZnO thin film antibacterial performance generally depends on surface morphology, film thickness, compositions, and crystalline quality. Thongsuriwong et al. investigated the influence of the thickness of ZnO film on their photocatalytic-activated antibacterial properties [120]. Deposition of ZnO NPs thin film on glass substrate was done by sol-gel dip-coating method. Three different thicknesses (218 nm, 312 nm, and 437 nm) were obtained, and the sample with the most negligible thickness has destroyed *E. coli* cells and inhibit growth under UV illumination up to 60 min, which might be attributed to increased surface roughness. The mechanism that may work on the ZnO thin film is direct interaction and generation of ROS rather than the release of  $Zn^{2+}$  ions. ZnO film with thickness less than 150 nm was successfully fabricated and coated on transparency glass by a similar approach, but with the assistance of polyvinylpyrrolidone (PVP) [122]. The fabricated coating demonstrated 90% transparency and anti-bactericidal properties against the gram-positive *S. aureus* ATCC 209P and gram-negative *E. coli* ATCC 25,922 bacteria both under natural light and in the darkness. A particle morphology modification in thin film beyond NPs such as nanorods [123, 124], quantum

nanodots [84], nanosheets [32, 125], nanocones [126], hierarchical structures [127–129], nanowires (NWs) [130] and so on may exhibit better antibacterial activity. However, further investigations are much needed to compare thin film properties with those of ZnO powders.

The relatively large bandgap of ZnO makes this material absorb only a small portion of sunlight, thus limiting the performance. To disclose the potentiality of ZnO NPs and extend its spectral absorption to the visible-light region, the doping approach with both anions and cations has become innovative, robust, and facile approaches [131]. A primary reason behind the choice of anions doping is that their  $2p$  orbital will hybridize with O  $2p$  orbital in the VB of ZnO, resulting in reduced bandgap and enhanced photo absorption ability [132, 133]. Fluorine-doped ZnO (ZnO:F) thin-film coated on glass through simplified spray pyrolysis was reported [121]. Thickness of film was linear to at.% of F dopant. It was found that F doping endowed simultaneous enhancement of optical transparency and antibacterial properties against *E. coli* and *Bacillus subtilis* bacteria. Specifically, F doping maintains film transparency of 80–90%, alters the bandgap structures, and contributes to free electron concentration in the system resulting in a significant amount of ROS production. Cations, usually in the form of metal cations, are also effective in reducing the band gap of ZnO, as they tend to occupy a transition state below the CB [39]. Hassan et al. reported antimicrobial thin film coating containing copper-doped ZnO. Cu is a well-established antibacterial material, yet it does not have sufficient transparency [134]. The high quality and easy-to-scale thin film with 90% optical transparency have been manufactured using an aerosol-assisted chemical vapor deposition (AACVD). The antibacterial activity against *E. coli* can be observed in dark and light environments with an optimum illumination of 18 h. In addition, the thin film decreased the WCA with increasing irradiation time. The antimicrobial mechanism was attributed to ROS oxidative stress agent production via photocatalytic and Fenton-type reactions and the release of metal ions. It should be noted that the photocatalytic-induced ROS generation did not take place in the dark. A recent report showed that Sn-doped ZnO NPs thin-film has broader antibacterial activities, not limited to *E. coli*, but also *S. aureus*, *K. pneumonia*, and *P. aeruginosa*.

In consideration of the necessity of thin-film coating with more functionalities, including self-cleaning, energy-saving, and solar energy conversion for application in intelligent windows, the combination with other functional materials, e.g., nanocomposite coating formation, is an exciting yet challenging approach [135]. This is because one can observe a trade-off when two dissimilar materials with different functionalities are combined. For example, incorporating ZnO NPs to VO<sub>2</sub> [136], an intelligent material with a thermochromic function, can improve the antibacterial property,

anti-corrosion, and biocompatibility of VO<sub>2</sub>. However, at the same time, it can degrade the switching efficiency ( $\Delta T$ , %) of VO<sub>2</sub>. In a summary, even though nanocomposite coating can add valuable functionalities, the appropriate parameters should be ensured to secure advantages from both components are included in nanocomposites. Moreover, it is troublesome to produce a coating with uniform-in-thickness, cost-effectiveness, and high homogeneity in a giant sheet of glass. One of the alternative solutions that can be attempted is making bioactive glasses containing ZnO NPs, because the incorporation of antibacterial active materials can be done during the glass manufacturing [137].

Building walls in hospitals and clinics is a habitable surface for bacteria and viruses, which must be cleaned regularly to reduce infection. Building walls consist of concrete and cement. ZnO NPs, as an antibacterial agent, can be coated onto it by incorporating them into paint suspension. Therefore, not only play a prominent role as an active antibacterial component, ZnO NPs may serve as anti-corrosion or self-cleaning agents [138–140]. It was proposed that ZnO NPs exhibited good activity against microorganisms at a pH value of 7, acceptable for use in the building [141]. In the form of nanofluid suspension, ZnO is still effective in restricting the growth of *E. coli* O157: H7 and *S. aureus* [142]. Similarly, acrylic paint containing a small amount of ZnO NPs showed promising results against gram-negative *P. aeruginosa* and Gram-positive *S. aureus* [139]. From these results, ZnO NPs incorporations into liquid paint suspension hold a promising prospect. Nevertheless, ZnO NPs can be directly mixed with cement, in which the antimicrobial properties can be activated under various illumination conditions (no light, natural light, and UV light) [143]. It is noteworthy that the antibacterial performance can further be enhanced by amine additive [139] or coating with silver [144]. While the hydroxyl group in amine may support the reactive oxygen production, and silver was oxidized to Ag<sup>+</sup> to co-agent in bacteria growth inhibition. Despite the full results, ZnO NPs is yet ready for deployment before one grand challenge is solved, that is the ZnO NPs antibacterial activity should remain active for years as they mix with the other ingredient of paints and sustain from environmental hazards [138, 145].

### ZnO NPs-based antibacterial coatings on papers

Due to fungal growth, the deterioration of paper-based documents in hospitals and research centers is of a primary concern, considering the damage it gives to the essential data, precious books, and rare manuscripts. In such way, the bacteria and viruses could be perched on the surface of those paper-based documents, transforming into infectious carriers for disease-causing bacteria and viruses. The remarkable and extreme care of documents using chemical methods and maintaining the temperature and humidity, which are proven

to limit the fungal and microorganism growth, becomes the old-century solution to date, although this approach requires repetitive implementation. The use of antibacterial coating on the paper could become an innovative approach to reduce the spread of infectious agents on the paper's surface. ZnO NPs has shown its excellent performance for environmental purification, self-cleaning, sensing, catalysis, and microorganism growth inhibition.

Over the past years, several methods, including physical and chemicals, have been used to prepare ZnO NPs coating on paper substrate. The main requirements of ZnO NPs coating for paper are high homogeneity, long-term use, and stability. Besides that, the highly desirable properties needed for ZnO coating on the paper are high transparency and flexibility, which would not alter the paper's original color and properties. Advanced development of ZnO coating on papers has been attempted to obtain excellent antibacterial properties of the paper. Ling's group has coated of ZnO NPs ( $d = 20$  nm) on the paper's surface using an ultrasound-assisted method, in which the ZnO-coated paper showed antibacterial activity against *E. coli*. Moreover, the combination of pigment slurry containing dispersant, China clay, and starch-stabilized ZnO has successfully produced ZnO nanoparticle-coated paper with pleasant properties for anti-fungal and UV-protection [146]. Moreover, adding some polymer such as carboxymethyl starch (CMS) to ZnO NPs enhanced the ISO-brightness and CIE whiteness of the coating compared to blank paper reference. The CMS is non-toxic and cheap additive used in food science [147]. The ZnO/CMS ( $25\text{--}50 \mu\text{g.mL}^{-1}$ ) showed the highest activity against *P. aeruginosa* (inhibition zone of  $5 \text{ mm} \pm 0.5$ ), while to be effective against *S. maltophilia*, the more amount of sample (above  $200 \mu\text{g.mL}^{-1}$ ) is required. Further investigation is needed to elucidate the effectiveness of the sample against bacteria according to the amount of CMS per nanoparticle in the ZnO/CMS samples.

The development of particle modification in ZnO thin film such as nanorods and NWs, although still a huge challenge, may exhibit better antibacterial activity on the papers,

where some studies showed that antimicrobial properties could be activated under visible light. The nanorods and NWs may add the surface roughness to the paper to prevent bacterial adhesion and act as a nanoscale knife to break the membrane of bacteria [148]. Chauhan and team found that ZnO NWs are antibacterial-active against *S. aureus*, *E. coli*, and *A. niger* [69]. The effectivity of ZnO NWs and nanorods-immobilized paper to reduce the growth of those microorganisms under visible light illumination could become very potential in many purposes such as medical paper in healthcare facilities, bacterial wipes, and food packaging. The thin-film coating with more specialized functionalities is obtained using combination with other materials to form nanocomposite has attracted the scientific community's attention. Afsharpour et al. have demonstrated that using the cellulosic nanocomposite coating of ZnO to the paper increased the durability of the papers undergo light aging and gave the glossiness on the paper, in addition to the increase of antibacterial activity, transparency, and no change in colors [149]. However, the production of ZnO with these morphologies or tandem materials using the present method is susceptible to agglomeration and retention, so several factors must be considered so that the agglomeration or retention does not happen, for example, the user of the *ex-situ* method to produce ZnO NWs coated on the paper.

To provide a clear insight into key requirements in achieving mass utilization of ZnO antibacterial coating on different substrate materials, we summarize main properties for ZnO coating to possess in Table 1.

## ZnO coating process and technological feasibility

Recently, the comprehensive research has been revealed to create uniform, reproducible, and reliable of thin-film layers to the substrate surface using several coating techniques. The ZnO thin films are conducted using various methods such as sol-gel, spray pyrolysis, hydrothermal method, atomic layer

**Table 1** Key performance indicator of ZnO antibacterial coating on various materials commonly used in healthcare facilities and medical devices

Substrates	Key performance						
	Hydro-phobicity	Mechanical stability	Flexi-bility	Reusa-bility	Bio-com-pati-bility	Trans-parency	Other
Textiles and fibers	✓		✓	✓			Washability
Metals and alloys	✓	✓			✓		Inertness to chemical
Polymers and plastics	✓	✓	✓		✓	✓	
Glasses and ceramics	✓					✓ (Glass)	
Papers	✓	✓	✓	✓			

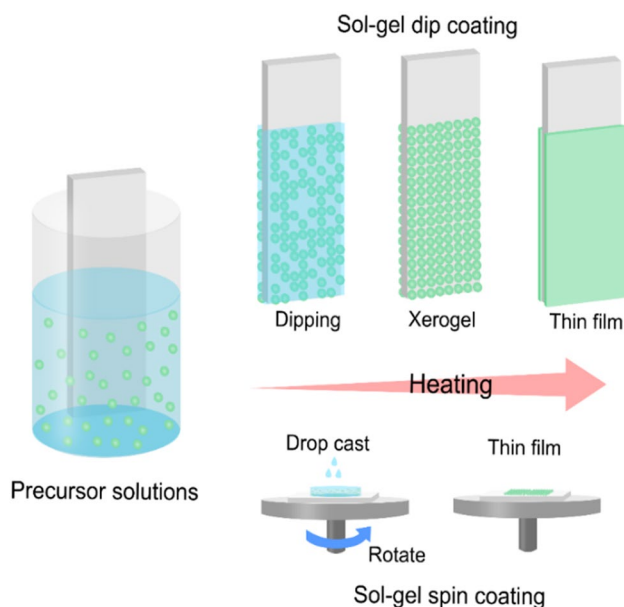
deposition, chemical vapor deposition, etc. There are many different performances and parameters resulted from different technique such as thickness, antibacterial performances, and bacterial adhesion properties. In this following section, we only summarize three general methods that mainly used to produce ZnO thin films.

### Sol-gel coating

The sol-gel technique is one of the most facile and effective methods for preparing a thin film coating containing porous, amorphous, or crystalline materials [150]. For many years, scientists have been using and developing this approach to be able to produce a high-quality thin film under room or moderate temperature [151]. The sol-gel coating process is illustrated in Fig. 5. The thickness of coating from sol-gel process are controllable by simply adjusting the reaction parameters such as sol concentration, dipping time, withdrawal rate, aging time, and post-treatment reaction, making the sol-gel approach an appropriate method for producing ZnO thin films with superior properties. In addition, a wide-choice substrates are available for sol-gel ZnO thin film coating which includes paper, metals, polymer, plastics, etc. that successfully employed in multifunctional smart coating. In the application of antimicrobial textile, the performance of ZnO coating such as high durability and long-term stability can be achieved by adjusting parameters. For instance, thermal curing steps become an important factor for ZnO antibacterial coating to the textiles which affects the cross-linked silica matrix onto the textile surface and also

affect the long-term stability of the antimicrobial functionality [152, 153]. Nanoparticle with rough textural properties during the curing facilitates to inactivate the microorganism attached on the textile. The interface of ZnO coating with the substrate is also a crucial factor to produce decent quality ZnO coating. Amakali et al. have synthesized ZnO thin film using sol-gel method which showed stable bonding between ZnO thin film and the micro glass substrate [154]. However, the mechanism of bonding between ZnO thin film and the micro-glass substrate in the molecular precursor still unclear. Therefore, further studies to elucidate the bonding mechanism are needed.

Biomedical coating for implants uses the sol-gel coating due to its simplicity, non-hazardous, cost-effective, rapid processing, and high-quality outcome [155]. The implant-related infection becomes a critical problem that needs to be prevented, considering the biofilm formation could destroy the tissue adjacent to the implant which can induce to poor vascularization, implant loosening, and dislocation [156]. Recent study showed that sol-gel coating technique can be used to coat metallic substrates containing monodispersed nanoparticles of Ag and Zn (sizes are in the range of 100–150 nm) with flower-like ZnO nanostructures. The rough surface texture of ZnO obtained from the sol-gel coating method displayed a superior hydrophobicity and liquid repellent properties to minimize biofilm formation [157]. The tribological and electrochemical behavior are another crucial factor in ZnO coating. In biomedical implant applications, the tribological properties of titanium is poor because of its low shear resistance in the oxide layer. The use of sol-gel ZnO coating improved their tribological properties and prevented wear phenomenon [158]. The corrosion resistance can be improved by nano compositing such as in the case of  $\text{Ag}_2\text{O}/\text{ZnO}/\text{NiO}$  thin film; however, future research can be devoted to gain deep understanding on the effect of coating thickness to the electrochemical behavior. In other works, using sol-gel method, Varshney et al. [159] reported the improvement of mechanical properties of hydroxyapatite/ $\text{MgO}/\text{ZnO}$  bio-ceramics for a bone-like material application. The utilization of this composite increases the mechanical properties of HA by enhancing its crystallinity and physical characteristics. Moreover, the chemical stability also can be achieved to a great extent which can be improved for bone regeneration materials. The superior properties in mechanical performance fits with the biomedical application.



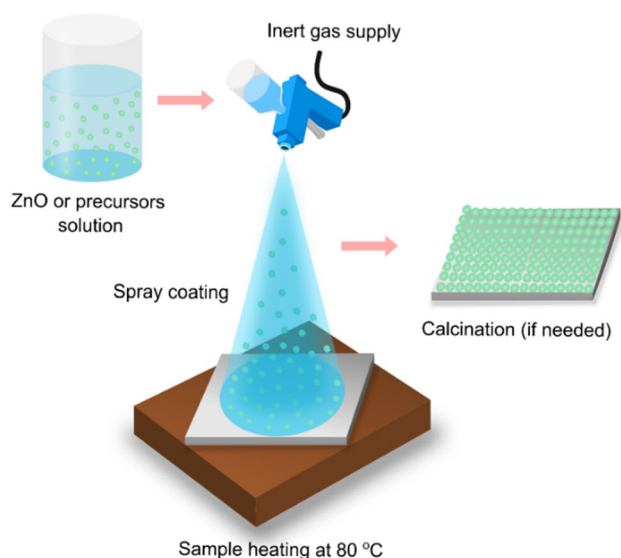
**Fig. 5** Sol-gel dip coating and spin coating processes of ZnO nanoparticles. Substrate representatives is glass

### Spray coating

Spray coating deposition method releases the liquid droplets through a nozzle onto a hot substrate assisted by flow of inert gas [160], forming a disk-like structure, where its shape and size rely upon the substrate temperature, volume, and momentum of sprayed droplets [161]. Spray coating

gathers much recent attention, because its economically and environmentally friendly, well-suited for both laboratory and large-scale production in industry, and minimal waste product compared to conventional processing techniques, for such as Radio Frequency (RF) magnetron sputtering, pulsed laser deposition (PLD), atomic layer deposition (ALD), chemical vapor deposition (CVD), and spin-coating [162]. Moreover, this method does not require a particular substrate specification and pretreatment which guarantee for homogeneous coating of ready-made applications such as construction and building application. The use of spray coating to produce thin film is promising as thickness and quality of the film are tailorable by a well-design preparative parameter. This technique can also be applied to form different thin film structures, such as layered, porous, and gradient composition thin film. Therefore, spray coating also become one of the best practices to coat ZnO thin film.

The spray coating process is illustrated in Fig. 6. The following spray coating process parameters greatly affect the spray-coated thin film; flow rate, temperature, nozzle-to-substrate distance, droplet size, precursor concentration, and deposition rate. According to Zargou et al., the optimum solution concentration at which the ZnO thin film showed optimum transparency and electrical conductivity is 15 mL. h<sup>-1</sup> [163]. Meanwhile, Alvers and co-workers reported that the microstructure and diameter of ZnO rods transform drastically with the increase of temperature increases [164]. As the nozzle-to-substrate distance get closer, the deposition area decreases. Therefore, the upper and lower limits of nozzle-to-substrate distance are generally determined based on the atomizer type and substrate size.

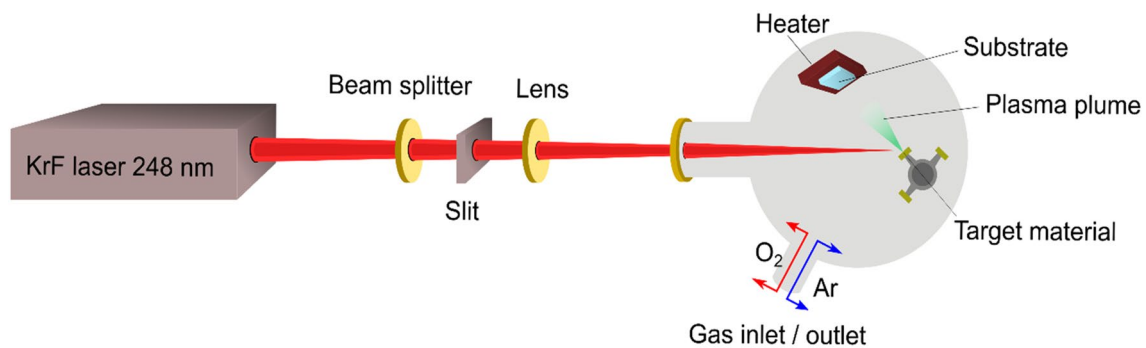


**Fig. 6** Schematic illustration of spray coating process of ZnO NPs

The use of spray coating process for depositing ZnO into a thin-film form has been applied in wide applications from optoelectronic devices to multifunctional coating. For instance, spray coating-deposited ZnO thin film plays as active layer of thin-film transistors (TFTs) [165]. Moreover, the spray coating process has not only been used to deposit pristine ZnO thin film, but also metal-doped ZnO which eventually improved the surface smoothness, structure compactness, and electrical conductivity of the thin film [166, 167]. Despite the benefits coming from its simple preparation, the spray coating process faces major challenges in facilitating the high yield thin-film production, the battling determination of growth temperature, and the possibility of sulfide oxidation under air atmosphere condition [168]. Owwoye et al. [169] investigated the effect of precursor concentration on ZnO thin films using the spray coating process and they found that the average crystal size of ZnO NPs depended on Zn molarities. Furthermore, micro-strain properties decreased as molarities increased, because the nanoparticle tends to agglomerate and subsequently form a single-crystal at high molarity. Hassan further revealed that the dislocation density ( $\delta$ ) is increased in the agglomerated ZnO thin film [170]. It can be concluded that the precursor concentration is essential for ZnO thin film properties as the relatively high concentration can cause the agglomeration which induce to a lower the mechanical properties of ZnO thin films. The future research should preferably be inclusive to overcome these issues.

### Pulsed laser deposition

Pulsed laser deposition (PLD) is one of physical methods to produce thin-film materials using vapor deposition process conducted under a vacuum condition. This process also utilizes molecular beam epitaxy and sputter deposition, where the pulsed laser is focused onto the target substrates. The plasma plumes resulting from laser pulse vaporization or small material removal under the process will allow the material flux for film growth. PLD is an attractive method to deposit and fabricate thin film growth of a complex and multi-compositions material. PLD is an unconventional method for large workpiece area; however, it is suitable for a laboratory scale application. This is because the spot size of pulsed laser is quite small, so it is only profitable to small target area even be less than 1 cm<sup>2</sup>. The pulsed laser deposition coating is illustrated in Fig. 7. As we know, there are so many applications of pulsed laser deposition method for examples biomedical, catalytic degradation, gas sensor, etc. These following factors have been proven to influence morphology, growth, and properties of the thin film fabricated by PLD: (i) laser parameters such as type of lasers, laser fluence, wavelength, pulse duration and repetition rate and (ii) the preparation conditions such as target-to-substrate



**Fig. 7** Schematic illustration of ZnO thin film fabrication by pulsed laser deposition (PLD) processes

distance, background gas and pressure, and substrate temperature) [171]. The ZnO thin film produced by a solid-state Nd:YAG laser ( $\lambda = 355$  nm) is thicker than that of thin film fabricated by KrF laser ( $\lambda = 248$  nm). Under this case, the thickness of ZnO thin films must be really considered by setting parameters involved in the pulsed laser deposition method.

The effect of substrate temperature on the structural and optical properties of Mg-doped ZnO thin films has been investigated in a literature [172]. Higher substrate temperature led to larger grain size, impacting greatly on the bandgap value. Recently, background gases and operating pressure affected to the thin film quality. The use of O<sub>2</sub>, Ar, and He as background gases with a pressure of 2.6 Pa has formed an amorphous film, while increasing pressure to 133.3 Pa has resulted in crystalline film formation [173]. The probable reason is related to their ion velocity, where the sequence of ion velocity reduction is O<sub>2</sub> > Ar > He.

The effect of annealing on the properties of In-doped ZnO thin films fabricated by PLD is recently investigated [174]. The crystallite size increased, because the thermal annealing improved crystal structure. Furthermore, the use of annealing at 500 °C under oxygen experienced not only an upward in the transmittance from 85 to 90% but also an increase in conductivity [174]. It can be concluded that substrate temperature, background gas, and heat treatment used in pulsed laser deposition were very influential on the grain size, crystalline structure, and film thickness of ZnO thin films.

Meanwhile, the effect of laser ablation time in producing ZnO NPs thin films was studied by Alamro et al. [175]. Enhancing the ablation time caused a higher amount of ZnO NPs concentration evidenced from UV–Vis results. Furthermore, the increment of ZnO NPs concentrations happened without being accompanied by any other elements, which confirmed the high purity of this synthetic method. Fu et al. focused on the effect of laser energy on the properties of Nd-doped indium zinc oxide thin films. The higher laser energy increases the target to radiate more particles which enhances the thickness of the ZnO film. Moreover, the increment of

laser energy causes the film to become denser in a particular range and increases the high optical transmittance by more than 80% [176]. Therefore, laser time and energy can affect ZnO NPs' thin film properties, such as the concentration and thickness of films that can be beneficial.

Table 2 compiles the vital benefits and drawbacks of different manufacturing technologies to fabricate ZnO thin film coating. Besides three major conventional coating technologies discuss above, we also list other coating deposition methods commonly used in lab-scale or industrial-scale processes.

## General critical parameters of the coating process

According to the above discussion, ZnO NPs can be deposited using multiple different coating methods. The approaches range in their capacity to manage particle packing density and layer thickness, their ability to employ various particles, and the method's complexity and instrumentation requirements. In general, ZnO NPs or any nanoparticles face the stability issue due to high surface energy, and thus they tend to stabilize themselves by lowering their surface energy via aggregation, coagulation, or agglomeration. However, the agglomeration NPs is undesirable for coating process, because particle packing density and layer thickness of the coating will be inhomogeneous. This is usually controlled by surface modification of NPs to minimize their surface energy and can be performed by attaching suitable ligand molecules on NPs surface to add solubility and inhibit agglomeration. NPs coating can be physically or chemically attached on the substrate aided by either covalent or hydrogen bonding and it can be in many forms, e.g., single, double, or multi-layered (depends on coating or deposition methods) [191]. In addition, the alteration in surface zeta potential of NPs can provide electrostatic repulsion and steric hindrance to NPs, preventing the potential of agglomeration [192]. NPs

**Table 2** Advantages and disadvantages of coating technology for ZnO thin-film applications

Coating technology for ZnO thin film fabrication	Advantages	Disadvantages	Substrate materials	Ref
Sol-gel and spin coating	Ease of manufacture Able to coat complex substrate Low processing temperature High purity	Excessive cost of raw materials Time consuming process	Glass, p-silicon, Ti metals, textile, polyester, indium tin oxide	[152, 153, 155, 158, 177–179]
Spray coating	No bulk particle melting Rapid and facile method	Hard to scale-up (yield is very low) Oxidation issue	Soda lime glass, AISI 4140 steel	[160, 162, 164, 166–168, 180, 181]
Pulsed laser deposition	Suitable for complex and multi-compositions material Controllable stoichiometric ratio for multicomponent coatings Facile preparation for variety of thin film materials	Low deposition rate and area Cost-extensive and complex	Si (100), glass, r-Plane sapphire	[172, 173, 182–184]
Atomic layer deposition	High material utilization efficiency Minimization of reaction volume	Non-uniform thickness Complex and tiring process	Silicon substrate, Ag nanowires	[185, 186]
Chemical vapor deposition	Flexible and facile method Suitable for complex structure The deposition rate is high Thick coatings can be readily obtained	High temperature operation By product harmful organics vapors	FTO (fluorine-doped tin oxide) glass, 304L stainless steel	[187, 188]
Plasma coating	Facile method with simple equipment A well-established coating process that is widely available and well understood	Difficult to control thickness High capital cost	AZ91 Mg alloy, Stainless steel	[189, 190]

coating process is affected by the quality of nanoparticles, hence by the parameters during the synthesis of nanoparticles. In wet chemical synthesis, these parameters are pH of the sol, additives used (such as capping agent, surfactant), the number of coating cycles, the effect of annealing temperature and calcination, among others. The main parameters of nanoparticles that influence the coating process are size, shape, and surface charge [193].

In physical coating method such as thermal spraying technology, molten or semi molten powders are deposited onto a substrate to produce a two-dimensional coating or in some cases, a three-dimensional self-standing material. The microstructure and properties of the material depend on the thermal and momentum characteristics of the impinging particulate, which are determined by both the spraying methodology and the type of feedstock materials employed [194]. Yang et al. studied sonication as the disruptive force to reform membranes around the surface of nanoparticles. The results showed that sonication amplitude, time, temperature, membrane ratio, sample volume, and density need to

be considered to optimize membrane coating of polymeric nanoparticles [195].

### Common failures in ZnO thin film coating

ZnO thin-film coating, despite the successful deposition process, will experience mechanical and chemical failures. Example of mechanical failures are thermal aging adhesive, cohesive, and fatigue failures, while chemical failures include depletion and defect failures [196]. The thermal aging failure will induce phase breakage due to heat exposure [197]. The evaluation of thermal aging is crucial, because the phase change will cause a reduction in strain resistance Young's modulus, and coating's volume [198]. The phenomenon of photodegradation caused by UV exposure frequently becomes a problem in ZnO coating and causes structural alteration due to the changed coating properties. The ZnO coating's durability against photooxidation has become a big challenge and is still being developed until



now [199]. Despite the coating durability failure, adhesion failures are also caused by the differences in the thermal expansion coefficient of ZnO-based films and substrate materials [200]. The multiple cracks which are commonly seen on the surface of ZnO films can provide separations between the films and the underneath material [201]. Therefore, it is crucial to investigate all kinds of failures to find the most effective approach to inhibit and overcome them.

Vu et al. [202] reported the mechanical properties of ZnO, SiO<sub>2</sub>, and Ag<sub>2</sub>O (ZnSiAg)-doped plasma-sprayed hydroxyapatite coating. The use of ZnSiAg-HA coated on Ti64 and cpTi samples showed that the coating had particularly cohesive failures as coating residue. In addition, some adhesive failures were also observed on the surface, but the delamination occurred from the top part at the epoxy resin site rather than delamination between the coating and substrate. However, the coating itself is still intact on the substrate representing strong bonding between the coating and the substrate. Furthermore, Ding et al. [203] studied about the properties of ZnO doped tantalum oxide (Ta<sub>x</sub>O<sub>y</sub>) multi-layer composite coatings on Ti6Al4V. The scratch test was done to examine the adhesion strength between the deposited coating and substrate. The utilization of the ZnO-Ta<sub>x</sub>O<sub>y</sub> sample showed less scratch width than the Ta<sub>x</sub>O<sub>y</sub> sample. This phenomenon happened because of the higher thickness of ZnO-Ta<sub>x</sub>O<sub>y</sub> than Ta<sub>x</sub>O<sub>y</sub>, which causes higher surface resistance to scratch. It can be concluded that adhesive and cohesive failure was seen on ZnO coating samples, where their performances were also affected by the thickness of coating and surface resistance to scratch.

Constant mechanical stress experienced by materials for a lengthy period can cause catastrophic failure at different time period. The durability is defined as material behavior to withstand destruction under long-term cyclic stress from external forces [204]. Fatigue is a process, where materials undergo repeated and continuous loading, which cause failure and decline of its mechanical performance [205]. Fatigue failure is usually caused by mechanical stresses such as mechanical impact, wave and current exposure, tensile loading (out-of-plane and shear displacements), where these situations will circumstantially encourage component failure through corrosion and cracking. In addition, the other reason for this failure might be due to the large temperature gap between the base and the top surface during processing such as quenching and annealing, which induce thermal stress at the interface between the binder and the top coating. This phenomenon can develop delamination and failure of ZnO coating. Fatigue failure becomes one of the problems we have to prioritize in the characterization of ZnO coated materials.

Kachoei et al. [206] revealed that ZnO nanocoating on NiTi orthodontic wires could prevent Ni release and fatigue effect and failure issues due to the better quality in surface

and improvement of corrosion properties of ZnO NPs. It has been proven that the ZnO nanocoating can supply a safer misapplication of NiTi devices and decrease failures caused by inadvertent piercing of tissues and bacterial invasion. Moreover, Wu et al. [207] developed ZnO nanocoating on the surface of polyether ether ketone (PEEK) powder using one-step hydrothermal method. The PEEK-based nanocomposite without addition of ZnO exhibited serious fatigue crack but the addition of 2.5% ZnO could remove serious fatigue crack and peeling. It can be known that the fatigue failure can't be separated from ZnO coating and due to the high use in medical application, clinical studies are needed to convince the effectivity of ZnO coating in vivo.

The last frequent failure in ZnO coating is depletion failure that usually comes with constant depletion of the coating because of its transformation to oxides, which eventually promote to the depletion of the coating phase [208]. The coating has some antioxidant phases in the coating which has a role as effective oxidation reservoirs for oxide development. Furthermore, the gradual depletion of the phase accelerates the degradation of chemical integrity of the coating [209]. Finally, the lack of phase reserve and the bond layer is completely depleted, the coating will lose its protection. Arukalam et al. [210] indicated the effect of hydrophobic poly (dimethylsiloxane)-ZnO coating using *P. aeruginosa* for marine application. The bacteria caused the primarily depletion which could be worse in the addition of immersion time in *P. aeruginosa* inoculum, the results showed that the coating's surface become more adhesive because of previous depletion of nano-roughness structure on the coating's surface. It can be concluded that bacteria could make depletion failure worse which is essential for marine application to prevent complete failure on the components.

## Future outlook

In realizing the relevant and mass applications, ZnO-based antibacterial active materials should be in the form of thin film coating. This review has summarized the antibacterial performance of ZnO coating on commonly found substrate materials in the biomedical and healthcare facilities, including metal alloys, polymers, plastics, ceramics, glasses, papers, textiles, and fibers. Herein we list the outlook of this emerging topic:

- The requirements of ZnO coating on each substrate material are different, given the fact that these materials are used for different purposes. For instance, on cotton and fabrics, ZnO coating must be chemically stable and still work effectively after multiple washing cycles with a non-ionic detergent at hospital laundry condition. On metals and alloys for implant materials,

ZnO coating should be biocompatible, in addition to the required properties such as high corrosion resistance, toughness, elasticity, low wear, and sufficient mechanical strength. While ZnO coating should have high hydrophobicity and flexibility upon the coating on polymers and plastics, ZnO coating on ceramic and glasses should possess high transparency, good interface compatibility with the substrate and good surface durability. On papers, ZnO coating should possess high homogeneity, long-term use, high stability, high transparency, and flexibility. Following the appropriate choice of substrates, deposition and thin film coating are also critical to support the use of ZnO as antibacterial coating.

- We are still lacking the exhaustive studies on clinical applications to investigate their side-toxicity and stability in exposed environment *in vitro* and *in vivo*. This prevents the development of ZnO-based antibacterial coating towards commercialization.
- It is expected that the antibacterial efficacy should not much decrease from the lab-scale experiments. To increase stability and biocompatibility, ZnO NPs may be functionalized by biomolecules such as carbohydrates and peptides, as these biomolecules have an equivalent property to human tissue. In addition, some of them act as co-antibacterial agents that can provide platform for bacterial killing.
- More emphasize on the structure–activity relationship (SAR) of photo-induced bacterial toxicity of ZnO should be given, especially for those in thin film coating form which is less pronounced than nanoparticulate forms.
- The global pandemic situation due to the spread of COVID-19 that remains high and rapid should encourage researchers to perform extensive studies on anti-virus coating performance of ZnO NPs. It is a critical to prevent the spread of viruses as earliest as possible before the mass infection. Thus, aside of the development of a high efficacy vaccine, this topic should be one of the important research directions of antimicroorganism functional coatings to combat global pandemic in the future.
- The development of multifunctional coating based on smart materials for multifunctional purposes such as energy generation and storage, environmental purification, and health quality improvement should be given more effort.
- The antibacterial mechanisms of ZnO thin film antibacterial coatings are not fully supported by a comprehensive investigation. Thus, further explanation of bacterial inhibition mechanism is needed, and future studies must be equipped with simulation modelling, theoretical experiments, and quantum chemistry cal-

culations. From this, we can understand the underlying mechanism of bacterial inactivation by ZnO which can be extended to various promising photoactive antibacterial materials.

## Summary and conclusions

In summary, this review purposes to highlight the importance of ZnO antibacterial coatings applications in medical devices and health care facilities. ZnO has given a significant contribution in the modern science and technology especially for biomedical application. It is among the most suitable materials in the field given the exciting physicochemical properties: good chemical stability, tuneable band structures, high optical transparency, excellent mechanical stability, surface hydrophobicity, low production cost, low toxicity, high biocompatibility, and excellent photocatalytic antibacterial inactivation. In the attempt to improve antimicrobial properties and the efficient utilization of ZnO, researchers have proposed many innovative approaches; morphological design, surface functionalization, heterostructure construction, and compositional tuning account for the most report. Furthermore, the comprehension of various antibacterial mechanisms of ZnO NPs and clashing perceptions among the researchers are discussed in this review. In a brief summary, the antimicrobial inactivation is classified into the following: (1) The  $Zn^{2+}$  ion is released and attached to the cell membrane, causing mechanical damage to the cell wall of microorganism. (2) The reactive oxygen species (ROS) generation acting as radical compounds and non-selective oxidants that damage most of biomolecules including lipids and amino acids, the two compounds that construct bacterial cell wall. (3) Direct attachment of ZnO NPs onto bacteria cell via electrostatic interaction also induces bacterial death due to corrosive nature of ZnO surface. Furthermore, ZnO NPs have wide compatibility with many substrate, and therefore, they can be coated onto various substrate from rigid to flexible and transparent substrates with almost negligible change in their antibacterial properties, demonstrating a promise for mass applications that are currently still limited. In this regards, some coating techniques ranging from physical and wet-chemical approaches may be suitable for the deposition of ZnO thin film. The physical approach offers excellent quality and controllable thickness of ZnO thin film, while wet-chemical coating provides more low-cost fabrication. Both processes have their own critical parameters. ZnO coating will also experience failures and damages for long term use such as thermal aging failure, adhesive failure, cohesive failure, fatigue failure, and depletion failure. Therefore, routine maintenance is needed to keep antibacterial properties stable. All in all, ZnO shows a bright promise as future

antibacterial coating materials due to the above-mentioned properties and technological feasibility.

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## Declarations

**Conflict of interest** There are no conflicts of interest to declare.

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