Visible-Light-Driven Photocatalytic Antimicrobial Mechanisms and Application Prospects

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Abstract

Due to the emergence of antibiotic resistance, the traditional antimicrobial approach to fight bacteria has become less effective and hence, interest in the light-driven physicochemical inactivation methods has grown. The photocatalytic systems based on semiconductors have demonstrated excellent antibacterial activity under visible light irradiation with high chemical stability, biocompatibility and efficient photogenerated charge carrier dynamics. This paper systematically explains the different mechanisms of photo-microbial inactivation such as generation of ROS, interfacial redox reactions, and membrane disruption pathway as well as surface engineering and thin film fabrication strategies. Comparative analysis of the coatings obtained by various physico-chemical methods (solution-derived hydrolysis-condensation routes and vapour deposition methods) shows that there are significant differences in the antimicrobial kinetics, structural stability, and photoresponse behaviour of these coatings. Experimental results have shown good inhibitory activity of this system against representative Gram-negative and Gram-positive bacteria, and the inhibitory activity has increased with the increase of the irradiation dose and the density of the catalytic surface. Despite the still unsolved issues such as the lack of understanding of photoactivated inorganic antibacterial systems' non-photocatalytic antibacterial effects and the potential difficulties in large-scale production, these systems offer tremendous potential for sustainable applications in biomedical engineering, public health infrastructure and environmental control.

Keywords

Photocatalytic inactivation, reactive oxygen species, charge carrier dynamics, sustainable applications, antimicrobial kinetics, irradiation dose, catalytic surface density.

1. Introduction

Microbial infections have become a critical issue across multiple sectors, including healthcare, food, and public health. Traditional antimicrobial methods (such as disinfectants and antibiotics) are becoming less effective due to factors such as increasing drug resistance and the potential toxicity and safety hazards it may bring to the environment and living organisms [1]. This has made inorganic antimicrobial materials a research hotspot. Among these antimicrobial materials, titanium dioxide holds particular promise.

Titanium dioxide (TiO_2) is a well-known photocatalytic material with advantages such as high chemical stability, non-toxicity, and low cost [2]. It is commonly used in environmental purification, self-cleaning materials, and antibacterial surfaces. Common crystal forms include anatase, rutile, and brookite [3]. Anatase TiO_2 has a higher reflectivity in the short-wavelength portion of visible light than rutile TiO_2, a bluish hue, and lower UV absorption than rutile, resulting in stronger photocatalytic activity and making it the most commonly used type in antibacterial applications [4]. As a photocatalyst, TiO_2 is activated by visible light to generate reactive oxygen species with strong oxidizing power, such as superoxide and hydroxyl radicals [5]. These reactive molecules can damage bacterial cell walls and membranes, leading to bacterial death [5]. Consequently, TiO_2 coatings exhibit sustained antibacterial activity when exposed to visible light, significantly inhibiting bacterial growth and spread.

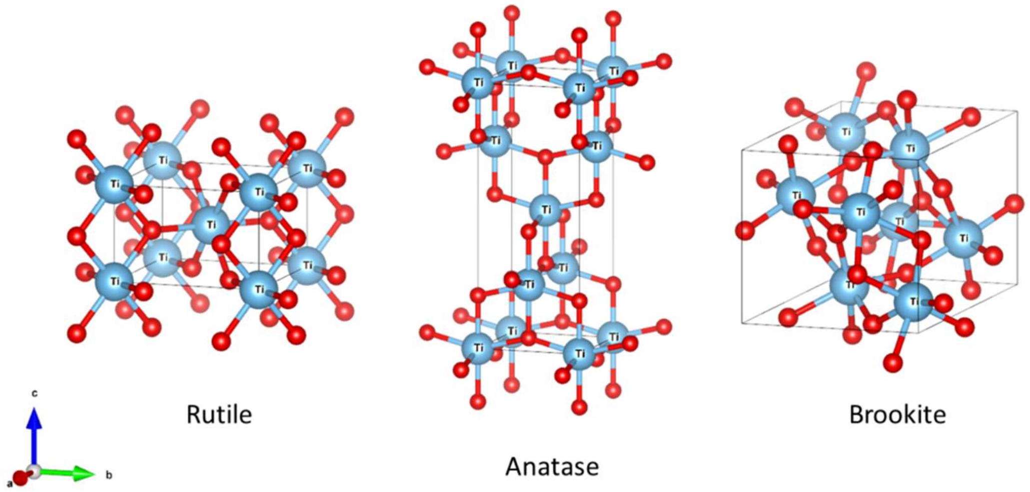

Figure 1. Three crystal structures of TiO_2 [3]

2. Preparation Method and Antibacterial Effect

Photocatalytic effect is the main reason why TiO_2 nanocoatings have antibacterial properties under visible light. They produce reactive oxygen species (ROS) under illumination which leads to the destruction of bacterial cell structures and a sterilizing effect [6]. The main applications they can be used for are self-cleaning, air purification and antibacterial coatings.

Various approaches have been followed for the preparation of nanoantibacterial coatings of TiO_2 . An approach is to prepare a solution of the TiO_2 nanoparticles using a sol-gel approach. An acidic aqueous solution, which contains glacial acetic acid and 37% hydrochloric acid, is added to the solution containing 97% titanium tetraisopropoxide (TTIP) to form hydrolysis and condensation reaction [7]. The stirring is continued at 60℃ for 90 minutes to obtain a TiO_2 nanoparticle solution [7]. This solution is used as a coating, giving any object self-cleaning and antimicrobial properties, such as masks. The antibacterial coating is done using different concentrations of the solutions of TiO_2 nanoparticles (0.5%, 1% and 2%) on a cloth mask [7]. The mask is then dried for about 24 hours. Antibacterial effect is then tested with the help of bacterial tests.

The antimicrobial activity of TiO_2 coated surface was found to be very effective, with good inhibition of common pathogens like Escherichia coli and Staphylococcus aureus at higher concentrations of TiO_2 , which was corroborated by the experiments conducted. Escherichia coli and *Staphylococcus aureus*.

The performance of the antimicrobial activity was tested by applying solutions of TiO_2 Nanoparticle at varying concentrations (0.5%, 1% and 2%) on cloth masks [7]. The results indicated that all the coatings were found to be effective in inhibiting the growth of *E. coli* and *Staphylococcus aureus*. The 2% TiO_2 coating in particular proved to be effective, resulting in a decrease of about 3.7 and 3.34 log units in the number of these bacteria after 18 hours [7]. The colony count of Escherichia coli (*E. coli*) reduced by 2.1 log cfu/cm^2 and the colony count of Staphylococcus aureus (S. aureus) reduced by 2.01 log cfu/cm^2 when 1% TiO_2 nanoparticle coating was performed [7]. The antibacterial activity of coating with 0.5% TiO_2 nanoparticles was about 0.5 log reduction of Escherichia coli colonies and 0.72 log reduction of Staphylococcus aureus colonies [7]. This means that the TiO_2 coating, even at a low concentration (0.5%) has a strong anti-bacterial effect, although not as pronounced as that of the 1% and 2% coatings. The untreated control group experienced almost no decrease in the numbers of bacteria, on the contrary. The TiO_2 nanocoating overall showed good antibacterial activity under visible light and the antibacterial activity of the nanocoating was found to increase with the increase of TiO_2 concentration, thus verifying the potential application of the nanocoating as an antibacterial material in protective equipment.

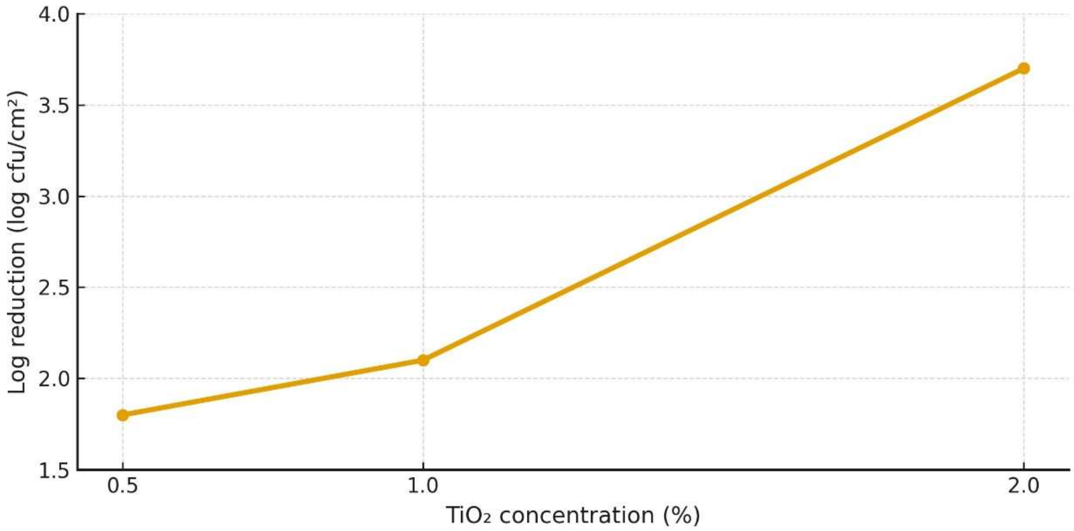

Figure 2. Relationship between TiO_2 concentration and antibacterial efficacy against E. coli, data were acquired from Ahmed et al. [7]

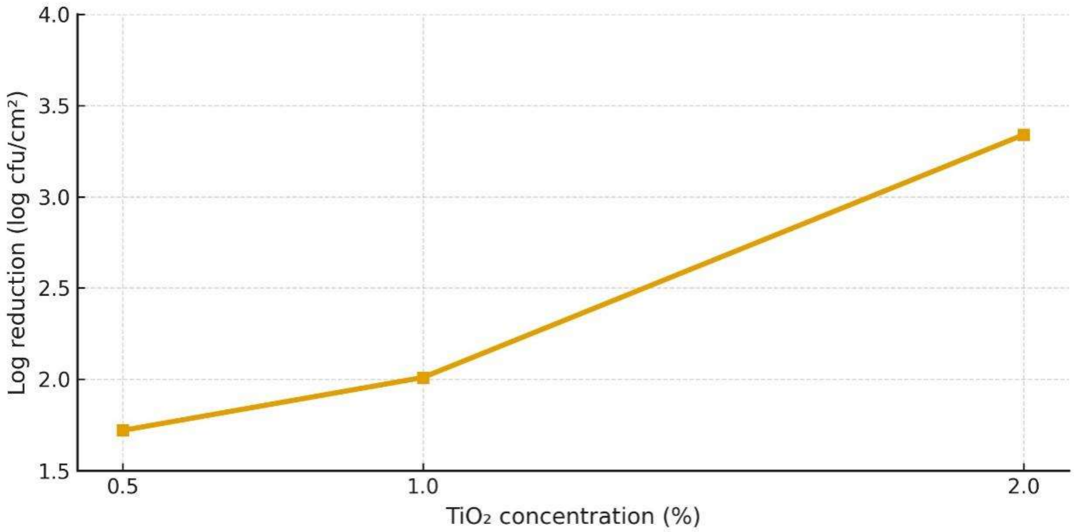

Figure 3. Relationship between TiO_2 concentration and antibacterial efficacy against *Staphylococcus aureus*, data were acquired from Ahmed et al. [7]

One approach is the pulsed pressure metal organic chemical vapor deposition (pp-MOCVD) [8]. Titanium tetraisopropoxide (TTIP) is used as the source of titanium and toluene as the solvent in this process. The coating is prepared by periodically injecting a liquid precursor, which evaporates at high temperatures to form the coating. After the deposition at 500°C on stainless steel or at 525°C on fused silicon substrates the thickness of the coating obtained is about 10 μm with a deposition rate of 15 nm/pulse [8]. This process is associated with a high vapor flux and pulsed injection, which results in a composite structure of anatase and rutile crystalline phases of the coating with co-deposition of amorphous carbon [8]. The addition of amorphous carbon increases the visible light absorption of the coating, and therefore its photocatalytic antimicrobial activity [8]. The coating fabricated by this method exhibits good adhesion and durability and shows high antimicrobial activity under the irradiation of visible light and can be used as an antimicrobial touch coating for public places, such as hospitals.

The TiO_2) nanocoating synthesized by this method exhibits a significant antibacterial activity especially under visible light irradiation. Such coating has demonstrated high level of *E. coli* inhibition (reduction) under visible light irradiation (at least 99.9% - a 3 to 4 log reduction) E.coli [8]. Such activity is attributed to co-deposition of amorphous carbon in the coating, which increases the range of light absorption by TiO_2 and consequently the photocatalytic activity of the coating under illumination in the visible range of the spectrum, as well as increases the adsorption ability of the coating's surface for organic matter, thus facilitating the generation of ROS, which can break the membrane of bacterial cells and cause their death [8]. Moreover, the coating also shows moderate antibacterial activity (a 2-log reduction) in the dark, which could be due to the after effect of ROS produced by the amorphous carbon and/or the influence of the coating on the cell membrane potential of bacteria [8].

3. Influencing Factors

The following is a list of factors that can affect the antimicrobial activity of the TiO_2 nanocoatings. The concentration of TiO_2 nanoparticle solution directly affects the antimicrobial effect. The higher concentrations of TiO_2 coatings have the stronger antimicrobial activity, and the lower concentrations have also a relatively weaker antimicrobial function [7]. The longer the exposure, the better the antimicrobial results. It was found that the number of bacterial colony-forming units (CFUs) decreased consistently with increasing contact time suggesting that the antimicrobial properties of the TiO_2 coated surfaces depend on time of exposure, particularly after 18 hours [7]. The coating absorbs visible light better, due to the increase in the light absorption range and co-deposition of amorphous carbon, which is an important factor in antimicrobial properties. The use of amorphous carbon makes it possible for the TiO_2 coatings to show an effective photocatalytic antimicrobial effect even under visible light, as TiO_2 is only highly reactive when exposed to UV-light. The nano-structure of the coating is an important factor in the antimicrobial function. In particular, the nanostructured anatase/rutile/carbon (NsARC) composite improves the photocatalytic efficiency, and hence the antibacterial effect [9]. This improvement is probably due to the greater surface area and the superhydrophilic surface of the coating, together with promoting good contact and degradation of the bacteria. During photocatalytic reaction with TiO_2 , a lot of ROS such as hydroxyl radicals and superoxide anions are produced and it may damage the cell membrane of bacteria to cause bacterial death [1]. The strength of the antibacterial properties depends on the ability and quantity of ROS generated. The efficacy of the NsARC coatings vary with different microorganisms. Although NsARC shows potent antibacterial activity against *E. coli*, *S. aureus*, *Pseudomonas aeruginosa*, and *Saccharomyces cerevisiae*, its antibacterial activity also is different under various lighting conditions [1]. For example, S. aureus exhibits a stronger antibacterial effect under both UV and visible light, while P. aeruginosa shows a stronger bactericidal effect under visible light.

4. Advantages

TiO_2 nanocoatings offer numerous advantages. Compared to silver-copper preparations, nano-TiO_2 can more thoroughly decompose bacterial cells, preventing secondary contamination from toxins released by dead bacteria [10]. TiO_2 coatings can continuously "regenerate" their bactericidal activity through light exposure, and even residual bacteria on the surface will be eliminated by a new round of light-excited free radicals, improving coating stability [11]. The bactericidal mechanism of TiO_2 primarily involves physical and chemical destruction, such as the generation of oxidative free radicals or direct structural damage to microbial cells. Unlike antibiotics, which act on specific molecular targets, this non-specific mode of action results in a significantly lower likelihood of inducing bacterial resistance. Currently, there are no reports of bacteria developing resistance to TiO_2 photocatalytic coatings.

5. Applications

TiO_2 antimicrobial coatings can be utilised on hospital door handles, operating room walls, and medical device surfaces, reducing the risk of cross-infection and creating a self-cleaning and self-sterilizing environment, helping to lower the spread of nosocomial infections [12]. In addition to its antimicrobial properties in healthcare and public health, TiO_2 nanocoatings are increasingly recognized for their potential in air pollution control. In urban environments, where PM2.5 pollution is becoming increasingly severe, TiO_2 coatings, due to their excellent photocatalytic degradation capabilities, have been widely used in air purification designs on building exteriors [13]. Japan has been a leader in the practical application and promotion of TiO_2 photocatalytic coating technology. According to reported cases, TiO_2 coatings were applied to selected buildings and urban structures in Tokyo as early as the early 2000s, demonstrating the initial adoption of this self-cleaning and air-purifying technology [14]. These coatings, typically anatase TiO_2 , are activated by sunlight and degrade airborne pollutants such as nitrogen oxides (NO_x), volatile organic compounds (VOCs), and fine particulate matter (PM2.5) [15]. These coatings, which are typically anatase TiO_2, are treated with self-cleaning technology. While maintaining a clean building appearance, they effectively adsorb and decompose pollutants adhering to the wall surface, producing harmless products such as water and carbon dioxide, thereby improving surrounding air quality. According to field test data from Japan's Ministry of the Environment and private organizations, under sufficient sunlight, each square meter of TiO_2 coating on certain treated building facades can degrade about 50 mg of NO_2 per day, which has practical benefits in alleviating the urban heat island effect and improving the air quality of the microenvironment [16]. Furthermore, the application of TiO_2-based coatings or composite films on the surface of medical implants, such as orthopedic devices, has demonstrated significant antibacterial efficacy while concurrently enhancing cell adhesion and overall biocompatibility. TiO_2 coatings containing Zn or Cu have a bactericidal rate exceeding 90% against *E. coli* and *S. aureus*, and can be used in biomedical devices such as orthopedic and dental implants [17]. Another type of SiO_2-TiO_2 core-shell nanocoating combines high strength with antimicrobial properties, showing broad application prospects in underwater antifouling for ships [18]. This coating can be used as an antifouling paint for underwater ship hulls, effectively inhibiting the growth of attached organisms and improving water flow efficiency due to its smooth surface [18]. The core-shell nanocoating is suitable for a variety of vehicles and structural surfaces, including ships, and offers multifunctional protection for substrates. With growing international demand for non-toxic, environmentally friendly antifouling coatings, TiO_2 nanocoatings, as a green and efficient antifouling material, have great industrial potential. Building materials such as glass, ceramics, tiles, and wall paints can be enhanced with TiO_2 nanocoatings to achieve multiple functions, including self-cleaning, antibacterial, and anti-fogging properties [19]. In high-traffic public areas such as subways, buses, schools, and office buildings, TiO_2 coatings can be applied to high-touch surfaces such as handrails, elevator buttons, and table and chair handles to effectively inhibit bacterial adhesion and growth, reducing the risk of cross-infection [6]. TiO_2 composite nanocoatings can significantly reduce E. coli counts on high-touch surfaces such as handrails in public places. In the food industry, they can form antimicrobial films on production lines and storage containers [20]. Moreover, they can also be used in food packaging processes to ensure food safety and sanitation [20].

6. Challenges

However, TiO_2 nanocoatings currently offer not all advantages, there are still numerous challenges remain. Previous studies have shown that nanostructured TiO_2 coatings, such as NsARC, exhibit antibacterial and biofilm-disrupting activity even in the absence of light, suggesting that their antimicrobial effect may involve non-photocatalytic mechanisms [9]. It may be due to free radicals released by carbon-based nanomaterials and cellular dehydration damage [9]. Further issues that need to be addressed include coating adhesion and durability. A big problem is the ability to firmly anchor TiO_2 nanocoatings onto a range of different types of substrates like metal, plastic and glass and also to maintain their wear and corrosion resistance over a long period of time [21]. Moreover, large scale production is accompanied with certain issues regarding scalability and safety [22]. Equipment and material costs for the production of high quality nanocoatings are high, and cost reduction and consistent performance are required for commercial production [22]. The nanofabrication and coating processes may be more expensive, but TiO_2 is a relatively low-cost material. Preparing a high-quality nano-TiO_2 material on a large scale and to be cost effective is the key to promotion [22]. While TiO_2 is considered biocompatible, there is still a need for evaluations of its release in the environment and bioaccumulation, especially in long-term exposure and human use [23].

7. Future Perpectives

In the future, further research should be done to explore the reaction pathways and free radical species of nanostructured TiO_2 in the dark state to design more effective all-weather antimicrobial coatings [24]. For practical application, better coating technologies may be required, like Pulsed Vacuum Deposition, Plasma Spraying and Magnetron sputtering to ensure good adhesion of the coating to the surface and uniformity of surface coverage [24]. It is also important to consider the potential effects of antimicrobial coatings on microbial communities and ecosystems, to prevent potential environmental risks. Regarding manufacturing processes, future development trends are toward low-energy consumption, green manufacturing, and scalability. Current research is exploring environmentally friendly processes such as hydrothermal synthesis, plasma-assisted deposition, and microwave-induced deposition, which can reduce the environmental impact of the production process while producing TiO_2 nanofilms with excellent properties at ambient temperature and pressure. Furthermore, large-scale continuous manufacturing methods based on 3D printing and roll-to-roll coating are also under development, promising the widespread application of antimicrobial coatings in medical devices, building components, and consumer electronics [25].

In addition, the intelligent upgrade of TiO_2 coatings has become one of the current research hotspots [26]. By combining TiO_2 with photoresponsive polymers, graphene quantum dots, phase change materials, etc., it is possible to achieve a sensing response to ambient light intensity, temperature, and even pollutant concentration, thereby regulating photocatalytic activity [26]. For example, certain responsive TiO_2 composite coatings can spontaneously increase ROS production when light intensity increases, and enter a low-energy consumption mode in low light or at night, taking into account both sterilization efficiency and energy utilization [27]. This type of "environmentally adaptive antibacterial coating" is particularly suitable for surface protection of medical devices and vehicles in variable lighting environments [27].

Even more cutting-edge is the integration of TiO_2 antimicrobial technology with emerging technologies such as artificial intelligence and big data analytics. In smart medical systems, surface "self-monitoring antimicrobial layers" based on sensors and data feedback are gradually entering the exploratory stage [6]. By integrating nano-TiO_2 coatings with biosensors, they can monitor bacterial loads and surface contamination in real time and intelligently determine whether light activation is needed, thereby achieving "on-demand sterilization" and optimizing resource utilization [6].

In general, the TiO_2 nanocoating technology is at a key moment of transition from basic research to multifunctional, multi-scenario, intelligent and green production. Future development should be directed towards not only improving its antibacterial characteristics but also combine it with other new material systems, new manufacturing technologies and digital control systems.

8. Conclusion

TiO_2 nanocoatings is evidenced under visible light which is mainly attributed to the production of reactive oxygen species through photocatalytic reactions. The preparation methods and structure of the coating greatly affect their antibacterial effects and applications. Despite these issues, including an unclear dark-state antibacterial mechanism, adhesion and cost control, the prospect of application for TiO_2 coatings is still wide due to their potential applications in the healthcare sector, public spaces and environmentally friendly materials. The stability and safety of their implementation in practical environment should be further investigated in the future for their industrialization.

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