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REVIEW

Biosynthesized Nanomaterials with Antioxidant and Antimicrobial Properties

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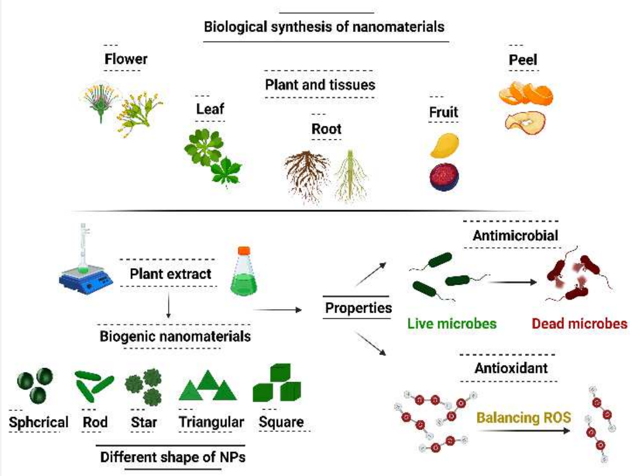
ABSTRACT

Nanomaterials are structures with dimensions less than 100 nm. Among different nanomaterials, metal- and carbon-based nanoarchitectures have attracted interest due to their ease of production, biocompatibility, low cost, excellent physio-chemistry characteristics, and biological activities. They are synthesized by various methods such as physical, chemical, and biological methods Biosynthesized nanomaterials exhibit remarkably improved biological activities such as antioxidant and antibacterial capabilities. Antioxidant nanomaterials can shield molecules from oxidation processes by decelerating or preventing them from oxidizing in the first place. These nanomaterials are widely used in the food industries and biomedical sectors. Several factors (e.g., size, shape, composition, and synthesized procedure) may influence the antimicrobial activity of these nanocompounds. It was shown that biosynthesized nanomaterials have higher antioxidant and antimicrobial activities than those by conventional methods. In the present review, we overview the antioxidant and antimicrobial activities of biosynthesized metal- and carbon-based nanoarchitectures. In addition, the mechanism of antimicrobial activity, as well as commonly used methods to measure the antioxidant activity of nanomaterials, are highlighted.



Keywords: Biosynthesized, metal- and carbon-based nanoarchitectures, antioxidant, antimicrobial

1. Introduction

Nanotechnology is a fascinating field of study that employs cutting-edge design, characterization, production, and application techniques to create structures on the nanoscale scale [1–3]. Nanosized materials, structures with a

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general size of 100 nm, can be categorized as organic (e.g., natural and synthetic polymers), and inorganic (such as metals and ceramic nanoparticles) [4]. Biosensing, medication delivery, and tissue engineering are the three primary disciplines of medicine that employ nanobiotechnology. Among them, metal- and carbon-based nano architectures have been attracting attention due to their simple synthesis approaches, biocompatibility, inexpensive, good physio-chemistry properties, and biological activities [5,6].

The production of safe and environmentally acceptable nanomaterials from biological sources such as plants, microbes, algae, and enzymes has piqued researchers' attention in recent years, given their easy synthesis technique and adaptability for a variety of applications. So, they can be made by physical, chemical, and green methods [5,7]. Among these routes, green synthesis owing to its easy synthesis, low-cost and eco-friendly is a proper method for the synthesis of metal- and carbon-based nanoarchitectures [8]. The green synthesis can be carried out by plant extract[8], microorganisms (bacteria and fungi) [9], and fruit juice [10]. The presence of various compounds such as proteins, polyphenols, and amino acids in natural compounds can be influenced by biological activities (e.g. antimicrobial activity, low toxicity, and antioxidant activity) of metal- and carbon-based nano architectures [11–13]. In this review, we descript the antioxidant and antimicrobial activities of biosynthesized metal- and carbon-based nanoarchitectures.

2. Nanomaterials with antioxidant activity

Antioxidants, also known as free-radical scavengers, are chemicals that protect molecules from oxidation by slowing or stopping oxidation [14]. The term "antioxidant" is generally used for two completely various groups of compounds such as industrial chemicals (synthetic polymers and nano-metals) and natural compounds (natural polymers and compounds) [15]. For example, plant-based antioxidants are types of natural compounds mostly used in the food industry [15]. Natural antioxidants are frequently restricted due to their great sensitivity to pH, light, oxygen, and other factors. Polyphenols in natural antioxidants also interact with a various of proteins, reducing their antioxidant action [16]. Endogenous antioxidants are antioxidants produced by the human body [17]. These compounds can prevent or reduce cell damage caused by free radicals, unstable molecules created by the body in response to environmental and other stresses. Oxidative stress can occur if the body is unable to remove reactive oxygen species (ROS) free radicals [17]. Naturally and artificial antioxidants are widely used in food and polymer industries [18]. According to the mechanism of action, they are categorized into different types including [19]:

Protective: These antioxidants are heterogeneous compounds consisting of metal chelating, sunscreens, hydroperoxide-decomposing agents, glutathione peroxidase, and superoxide dismutase enzymes. They are compelled to reduce the rate of initiation [19,20].

Chain-breaking: These chemicals function as radical traps, slowing or stopping the autoxidation cycle by competing with the propagation reactions. For example, the reaction between the antioxidant material (AH) and a $ROO^{\bullet}$ to produce a ROOH and $A^{\bullet}$, which traps a second $ROO^{\bullet}$ producing neutral end products [21].

Nanoantioxidants: They are nanomaterials that have resulted in a decrease in the rate of autoxidation and/or a reduction in the commencement procedures. Nanoantioxidants have longer stability than tiny molecules like vitamin E and b-carotene, allowing them to bypass rapid metabolism and target specific areas. Nanomaterials such as Ag nanoparticles (Ag NPs), and ZnO nanoparticles (ZnO NPs) could be used as carriers to carry antioxidant compounds or could have inherent antioxidant properties [22].

2.1. Measuring antioxidant activity

Antioxidant capacity can be measured in seven different ways. These methods work by interacting directly with reactive chemicals or free radicals reacting with metal ions [23].

***Oxygen radical absorbance capacity* (ORAC):** This method determines the efficiency of different natural antioxidants, existent in plasma/tissue, in inhibiting the $ROO^{\bullet}$ oxidation of the fluorescein. ORAC is based on the inhibition of $ROO^{\bullet}$ oxidation through antioxidants, which can be distinguished as loss in the fluorescence intensity during $ROO^{\bullet}$ damage. This method is not appropriate for determining a single antioxidant. Azo based-compounds, e.g., 2,2′-azobis(2-amidinopropane) dihydrochloride (AAPH) are employed as a $ROO^{\bullet}$ generator. ORAC technique was restricted to hydrophilic antioxidants due to the aqueous environment [24].

***Ferric reducing antioxidant power* (FRAP):** This technique determines the power of antioxidants with the assistance of an oxidant, i.e., ferric ions (Fe^{3+}). The values of FRAP are achieved by comparing the absorbance variation at 593 nm in examination reaction mixtures with those including Fe^{2+} in defined concentration. In this approach, antioxidants contained in Fe(III)/tripyridyltriazine in stoichiometric excess reduce materials to create a blue ferrous. Indeed, the absorbance change is related to the combined Fe^{3+} reducing/antioxidant that determined the power of the antioxidants in the sample [25].

Trolox equivalent antioxidant capacity* (TEAC)*: When 2,2′-azinobis-(3-ethylbenzothiazoline-6-sulfonic acid) (ABTS) is incubated in the existence of peroxidase and H_2O_2 or the existence of $^{\bullet}OH$, $ROO^{\bullet}$, $RO^{\bullet}$ and inorganic radicals, $ABTS^{\bullet +}$ is produced. Once antioxidants are added before the H_2O_2 addition, the antioxidants hunt the radicals produced by the H_2O_2, delaying the $ABTS^{\bullet +}$ formation, thus inducing an increase in the inhibition absorbance percentage. This method is based on the inhibition through antioxidants of the $ABTS^{\bullet +}$ absorbance, which has a specific long-wavelength absorption displaying maxima at 660, 734, and 820 nm [26].

***Trapping antioxidant parameter* (TRAP):** This method is based on the determination of O_2 consumption during a lipid peroxidation reaction produced through azo-compound decomposition such as AAPH. TRAP is the most extensively used technique for determining plasma/serum antioxidant capacity. Once AAPH is added to the plasma, the oxidation of the oxidizable materials is observed by determining the O_2 consumed during the reaction.

Dichlorofluorescin-diacetate* (DCFH-DA)** [27]:*** The AAPH is used to form peroxyl radicals, and DCFH-DA is used as the oxidizable compound for the peroxyl radicals in this process. The DCFH-DA oxidation led to dichlorofluorescein (DCF). The formed DCF can be observed in either spectrophotometric or fluorometric instruments [28].

Total oxidant scavenging capacity* (TOSC)** [29]:*** With the assistance of this method, the absorbance capacity values of antioxidants towards three strong oxidants (e.g., $^{\bullet}OH$, $ROO^{\bullet}$ and $ONOO^{-}$) can be measured. These oxidants were produced through the reaction of iron plus ascorbate-driven Fenton, AAPH thermal homolysis, and 3-morpholinosydnonimine *N*-ethylcarbamide, respectively. They react with KMBA (alpha-keto-gamma-methiolbutyric acid), which is then oxidized to create ethylene. The ability of the materials to suppress ethylene production compared with a control reaction is used to determine their antioxidant activity.

***2,2-diphenyl-1picrylhydrazyl* (DPPH•):** DPPH• is a stable free radical, because of the electron delocalization on the whole structure. As a result, unlike most free radicals, it does not dimerize. The delocalization on the DPPH• molecule defines the happening of a purple color, with a maximum absorption peak around 517 nm. The DPPHH is formed when DPPH• combines with a hydrogen donor, and the violet hue disappears. The decrease in absorbance is precisely proportional to the antioxidant content. The antioxidant capacity of materials in DPPH• solution was determined using the spectrophotometric approach. Either of the above methods can be used to determine the dependability of tested antioxidant compounds [30, 31].

2.2. Antioxidant nanomaterials

Numerous types of nanomaterials have inherent antioxidant characteristics that are not influenced by antioxidant functionalization, but rather depend on the material's surface features. Inorganic metal nanoparticles, especially those synthesized by the green method are the most widely inherent antioxidant nanomaterials [32]. However, antioxidant nanomaterials include organic nanoparticles with intrinsic antioxidant capabilities, such as conductive polymers (polyaniline and polypyrrole) and antioxidant-modified organic nanoparticles [33–35]. Recent research has shown that greenly manufactured metal- and carbon-based nanocompounds have stronger antioxidant activity than chemically synthesized nanocompounds. Plants' antioxidant activity is influenced by the presence of proteins, polyphenols, and amino acids as reducing agents in metal- and carbon-based nanoarchitectures. For instance, TiO_2 nanoparticles synthesized by *Psidium guajava* extract showed higher antioxidant activity than the conventionally synthesized counterpart. Apparently, the presence of phenolic compounds in the produced TiO_2 nanoparticles as decorating and capping agents increased the antioxidant activity [36]. This action of metal nanoparticles has important implications for medical applications, particularly in tissues that are suffering from oxidative stress [33].

Fruit juice (grape, orange, and lime), potato, and plant leaves have all been used in the green production of carbon nanodots (CNDs) [37–42]. Coriander leaves, for example, have been shown to produce antioxidant multifunctional

CNDs for sensors and bioimaging applications (**Figure 1A**). CNDs' antioxidant activity was dose-dependent, meaning that as the quantity of CNDs grew, so did the amount of radical scavenging activity (**Figures 1B** and **C**) [43].

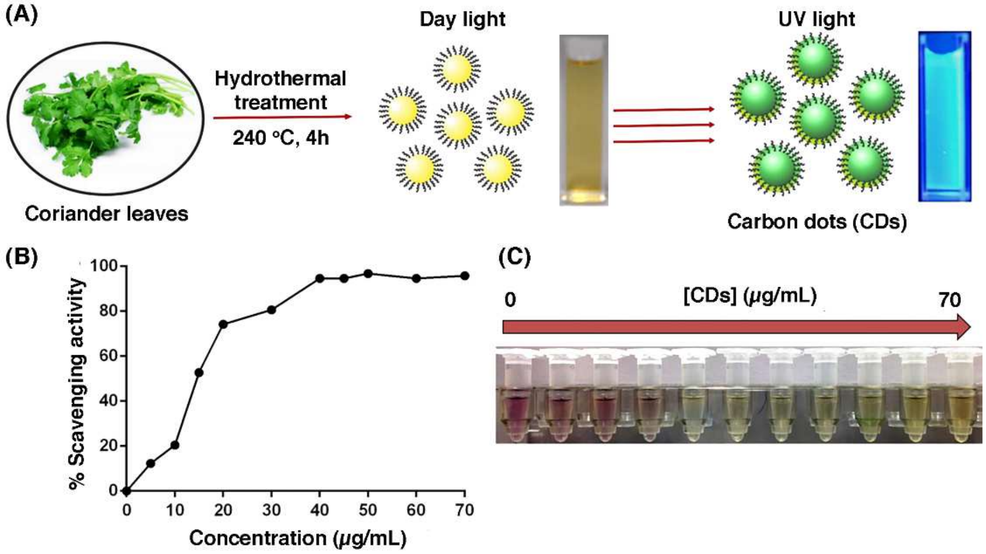

Figure 1. (**A**) Representation illustration describing the green synthesis of CNDs from coriander leaves as follows: chopped coriander leaves (5 g) dissolved in distilled water (40 mL) and then underwent hydrothermal treatment for 4 h at 240 °C. Finally, the solution was allowed to cool naturally and CNDs obtained by filtration through a filter membrane. (**B**) 2,2-diphenyl-1-picrylhydrazyl (DPPH) free radical scavenging activity of CNDs in methanolic DPPH solution. (**C**) Photographic image of decolorizing of DPPH solution with a progressive increase in CNDs concentration. Reprinted with permission from [43].

Antioxidant ZnO nanoparticles are fabricated by Eucalyptus globulus as reducing and capping agents for photocatalytic applications [44]. When compared to those made by conventional chemical procedures, the capping of natural chemicals on the ZnO surface and lower particle size can boost their antioxidant activity. Cuminic aldehyde and -Sitosterol in Eucalyptus globulus had a significant impact on the antioxidant activity of ZnO NPs (**Figure 2**) [44].

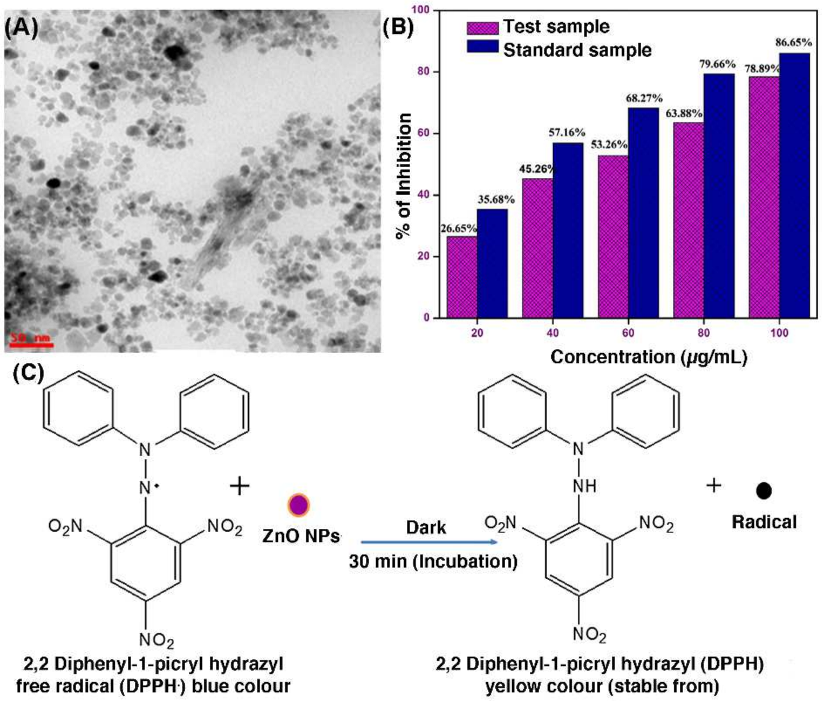

Figure 2. (**A**) TEM image of ZnO NPs at magnification 50 nm, (**B**) Antioxidant activity of various concentrations of synthesized ZnO NPs and standard ascorbic acid by DPPH radical scavenger. (**C**) Mechanism of interaction between DPPH radicle scavenger and ZnO NPs which formed a stable DPPH molecule. Reprinted with permission from [44].

Biogenic synthesis of silver nanoparticles (Ag NPs) by fruit juice and extract, coleus vettiveroids, nothapodytes nimmoniana, peel extract of ananas comosus, garcinia mangostana, Elephantopus scaber leaf extract, Averrhoa carambola, ornamental plant, Lantana Camara, apple extract, Emblica Officinalis, Prosopis fractal, etc. is reported by many research groups [32, 45–53]. In one study, AgNPs with biological activities were synthesized by the Ananas comosus peel extracted for biomedical applications. The functional group existing in ananas comosus e.g., flavonoids and phenolic acids were responsible for the Ag(I) reduction and the enhancement of AgNPs biological activities such as antioxidant and antimicrobial [49].

Bond creation between nanoparticles and natural antioxidants is another way of generating antioxidant nanomaterials, in addition to the methods outlined above. The coupling technique, for example, was used to create silica nanospheres covalently coupled to the caffeic acid antioxidant (SiCA) (**Figures 3A** and **B**). The radical-scavenging activity of SiCA was lower than the CA; though, singlet oxygen quenching of SiCA was enhanced. In addition, CA was shielded against protein binding, suggesting that SiCA is more likely to maintain CA's antioxidant action in biological media (**Figure 3C**) [16].

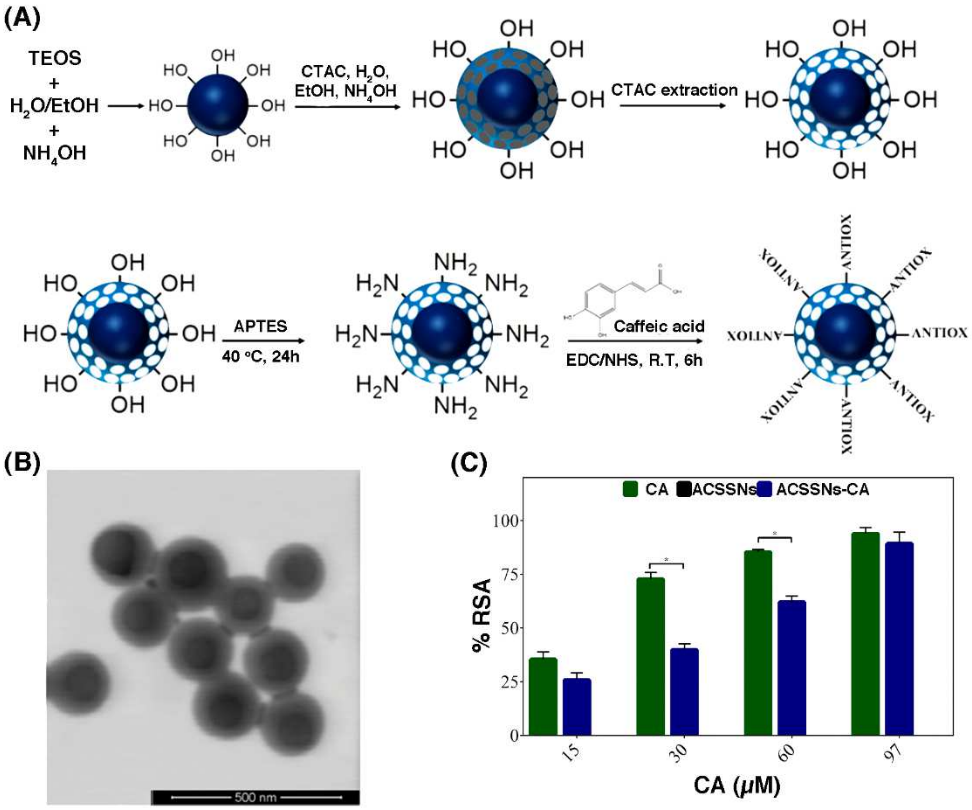

Figure 3. (**A**) Procedure scheme of the core-shell silica nanospheres with immobilized caffeic acid (SiCA). (**B**) TEM image of core-shell SiCA with magnification 500 nm (**C**) Radical-scavenging activity (%RSA) of caffeic acid (CA) and core-shell SiCA with radical DPPH•. A comparison of the %RSA between CA and SiCA displayed that the antioxidant activity was directly associated with the %CA on the silica nanoparticles. Reprinted with permission from [16].

3. Antimicrobial activity of nanomaterials

Pathogenic bacteria expose humans, resulting in a slew of pathogen-related infections and diseases every year [4]. Microbial infections are the main cause of prolonged infections and death. Antibiotics have been the favored treatment technique for microbial infections owing to their cost-effectiveness and powerful consequences. Most antibiotic resistance mechanisms, on the other hand, are irrelevant for nanomaterials since their action mode involves direct contact with the bacterial cell wall, without the need to infiltrate the cell. This increases the prospect that nanomaterials will have a lower proclivity for generating bacterial resistance than antibiotics. As a result, new and interesting nanomaterials with antibacterial activity have been given special study [54].

Inherently antimicrobial nanoparticles (e.g. Ag NPs and ZnO NPs), carriers-based antimicrobial nanomaterials, and nanomaterials containing both functional characteristics are the three types of antimicrobial nanomaterials. Furthermore, as compared to chemically synthesized nanomaterials, green-synthesized nanomaterials had greater antimicrobial activity [55–57].

3.1. Mechanisms of antimicrobial activity

Microorganisms develop resistance to antibiotics through a variety of processes [33, 57]. In this regard, eight mechanisms are proposed: [4]

I) Antibiotics activated efflux from bacteria by upregulation of efflux pumps.
II) Obtaining alternate metabolic pathways to those that are hampered by the medication.
III) Bacterial cell wall permeability is reduced, limiting antimicrobial availability in target areas.
IV) Antibiotic degradation
V) Antibiotic enzymatic modification
VI) Antibiotic goals modification

VII) Goal enzyme overproduction

Antimicrobial resistance genes are transferred within biofilm members via quorum detection.

Nanomaterials having antibacterial properties suppress or kill microorganisms by one or more methods. Nanomaterials' antibacterial mechanisms are related to their application (**Figure 4**) [4].

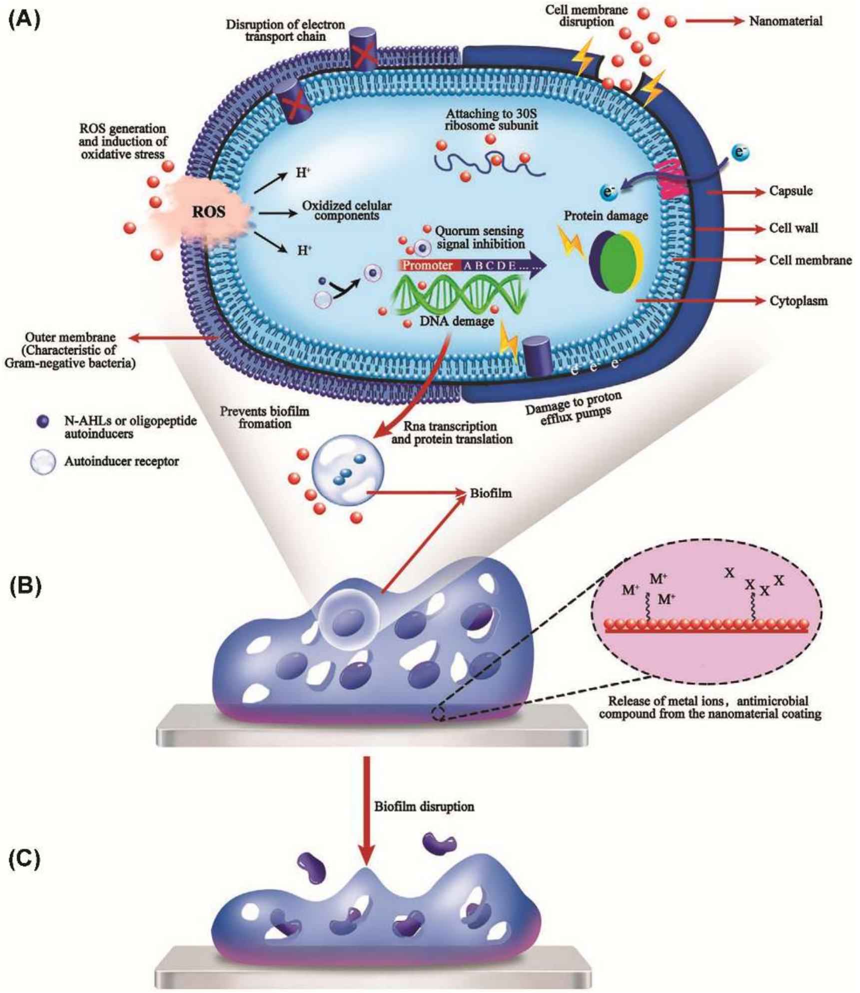

Figure 4. (**A**) Reasonable antimicrobial mechanisms of nanomaterials at various parts of a bacterium. (**B**) Drawing of the biofilm formation inhibition on surfaces covered by nanomaterials. (**C**) The double functions of nanomaterials in killing bacteria and inhibiting biofilm consequence in biofilm disruption. Reprinted with permission from [4].

3.2. Antimicrobial nanomaterials

Currently, biogenic nanotechnology-based methods have been accepted as eco-friendly and cost-effective methods with numerous biomedical applications [12, 13, 58]. The method has achieved considerable attention as a reliable and environmental-friendly procedure for synthesizing an extensive range of nanomaterials including metal and metal oxide nanoparticles, hybrid nanomaterials, carbon nano-dots, and bioinspired materials [59]. Indeed, the biological components such as terpenoids, flavonoids, amides, alkaloids, and aldehydes as reducing agents and solvent systems play a key role in the sustainability, biocompatibility, and low toxicity of nanomaterials [60–62]. For example, biogenically synthesized nanomaterials are suitably decorated with natural flavonoids that suppress enzymatic activity. This approach has prevented the production of nucleic acids in a variety of microorganisms, and it may now be used safely in a variety of medicinal applications [63]. Furthermore, natural chemicals have been used to increase the biological activities of nanomaterials connected with surface engineering approaches [59]. n compared to chemically synthesized Ni NPs, biogenic-synthesized Ni NPs exhibited greater antibacterial efficacy against the majority of the microorganisms examined [55]. In general, the antimicrobial efficiency of nanomaterials is determined by several parameters, including the material used to synthesize the nanoparticles, as well as the shape and particle size of the nanomaterials [64]. It was shown that nanomaterials with the size of > 20 nm can penetrate the cell wall of bacteria and in turn prevent the biochemical ways via cell organelles damage which eventually cause to death of bacteria [65]. Metal nanomaterials such as CuO (<30nm), ZnO (<20 nm), and Fe_2O_3 (<35nm) with different compositions and size particles in spherical shape showed various microbicidal activity against microbes [66]. It was observed that their antibacterial activity was in the order of ZnO > CuO > Fe_2O_3 [66]. In contrast to spherical/rod-shaped Ag NPs, triangular-shaped Ag NPs showed a significantly higher antibacterial potential [67].

In some studies, ZnO NPs exhibited higher antibacterial activity against S. *aureus* in comparison to CeO_2, TiO_2, MgO, CuO, and Al_2O_3 nanoparticles. Indeed, the greater interaction between ZnO NPs and the bacterial cell wall, compared with other metal oxides, led to the disruption of cell integrity [68].

The shape of biosynthesized metal nanostructures has also an important effect on antimicrobial activity. For example, biosynthesized ZnO NPs with different shapes such as nanorod, nanosphere, nanoflower, nanowire, nanobelt, and nanoflake exposed higher antibacterial activity against Gram-negative and Gram-positive bacteria than those NPs synthesized through conventional chemical routes [69].

Biogenic Pd, Se, Ce, and Te NPs have also revealed antimicrobial activity [70]. Pd NPs synthesized by using *M. oleifera* peel extract and exanimated for antimicrobial activity against *E. coli* and *S. aureus* [71]. The antimicrobial study showed that the Pd NPs efficiently repressed the growth of *E. coli* and *S. aureus* strains. In the same research biosynthesized CeO_2 NPs from *M. oleifera* peel extract and investigated their property against the *E. coli* and *S. aureus* strains [72]. An antibacterial examination verified the effective inhibition of *E. coli* and *S. aureus* strains. Rod-shaped Te NPs synthesized by using *Bacillus sp. BZ.* The Te NPs presented antibacterial activity against various strains such as *K. pneumonia, S. Typhi*, *P. aeruginosa*, and *S. aureus* [73].

Carbon nanodots (CNDs), also known as carbon quantum dots, are tiny carbon particles with a diameter of fewer than 10 nanometers that have emerged as a very promising new platform for visible/natural light-activated microbicidal compounds [74][75]. Biosynthesized CNDs derived from natural sources possess antimicrobial activities. They are utilized to improve the antibacterial properties of other antimicrobial materials. CNDs have been produced from sago starch to boost the antibacterial properties of poly (amidoamine)[76].

The production of reactive oxygen species (ROS) is most likely the major cause of CNDs' antibacterial activity. Photoexcited CNDs can produce ROS, which is known to kill/hinder microbes. As represented in **Figure 5**, CNDs adherence to bacterial surfaces, photoinduced production of ROS, penetration and breakdown of the bacterial cell wall and/or membrane, and the onset of oxidative stress with DNA/RNA damages are all part of the antibacterial action mechanism [77–79]. CNDs engage with bacteria cells under light irradiation and efficiently create ROS by activating O_2 in the air/water [80,81].

3.3. Innovative nanoemulsions

Nanoemulsions are dispersions of nano drops of two immiscible liquids that can avoid coalescence phenomena thanks to the Brownian motion and stabilization fabrication methodology as homogenization, microfluidization, and sonication [82,83]. The most innovative nanoemulsions are typically classified into oil-in-water (O/W) or water-in-oil (W/O) types. This kind of nanotechnology usually reports high loading efficiency for lipophilic molecules such as vitamins, natural antimicrobials, or high bioavailability [84]. They also took the use of chitosan and its derivatives' potential to transport anti-inflammatory molecules including Coenzyme Q10, lycopene, curcumin, and essential oils with antimicrobial qualities (data under processing) [85].

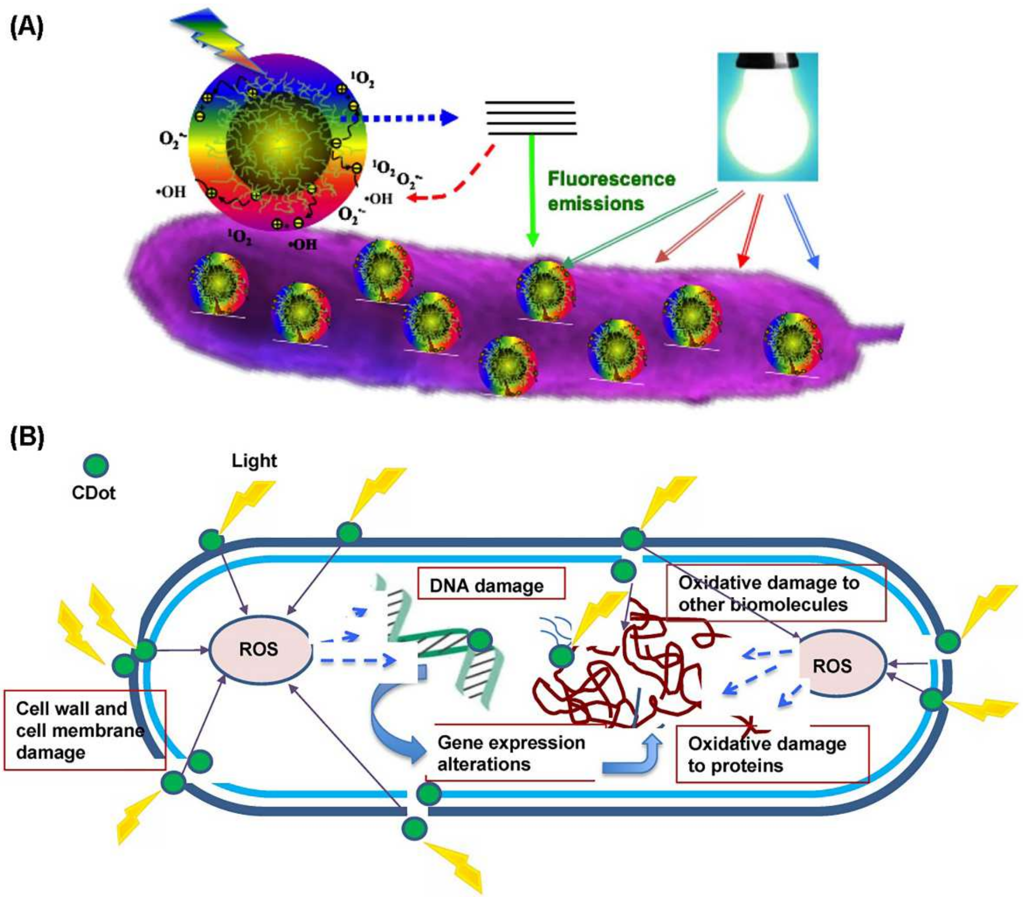

Figure 5. Schematic diagram of the antibacterial mechanism for CNDs. (**A**) CNDs adhesion to bacterial, and light-induced generation of ROS. Reprinted with permission from [86], (**B**) Intracellular ROS led to bacterial cell damage. Reprinted with permission from [74].

4. Challenges and future perspective

Although the synthesis of nanomaterials using bio-sources is simple, it poses a difficulty in terms of achieving well-defined, active, and stable NPs for a variety of applications. As a result, throughout the production process, the following issues should be considered: (i) natural resource/material selection, (ii) stability, (iii) activity, and (iv) repeatability.

Because biosynthesized nanomaterials have been identified as possessing antioxidant and antibacterial properties, scientists and engineers should now evaluate methods for mass-producing these nanomaterials. This saves not just money but also the environment by preserving natural resources, lowering energy usage, and contributing to the betterment of human existence on the planet. To build biosynthesized smart nanosystems on a big scale and save billions of lives worldwide from life-threatening illnesses, a multidisciplinary partnership between academic research and the pharmaceutical sector will be critical.

5.Conclusions

Biosynthesized nanomaterials are widely recognized as synthetic biological agents due to the presence of diverse components such as proteins, polyphenols, and amino acids in natural molecules that might impact biological activities (e.g. antimicrobial activity, low toxicity, and antioxidant activity). These nanomaterials with antioxidant and antimicrobial activities have attracted great interest in biomedical applications because of their high biocompatibility and low toxicity. Nanoantioxidants are nanomaterials that have slowed the rate of autoxidation and/or reduced the initiation processes. For measuring the antioxidant activity of nanomaterials, seven procedures have been proposed, including oxygen radical absorbance capacity, ferric reducing antioxidant power, Trolox equivalent antioxidant capacity, trapping antioxidant parameter, dichlorofluorescein-diacetate, total oxidant scavenging capacity, and 2,2-diphenyl-1picrylhydrazyl. Antimicrobial biosynthesized nanomaterials are mostly utilized in various biomedical applications. Their antimicrobial activity depended on size, shape, composition, and synthesized method. The antibacterial activity of nanoemulsions made using a green approach is greater than that of nanoemulsions made using traditional methods.

Authors' contributions

All authors contributed to drafting and revising of the paper and agreed to be responsible for all the aspects of this work.

Declaration of competing interest

The authors declare no competing interest.

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