Low-dimensional nanomaterials for antibacterial applications

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The excessive use of antibiotics has led to a rise in drug-resistant bacteria. These “superbugs” are continuously emerging and becoming increasingly harder to treat. As a result, new and effective treatment protocols that have minimal risks of generating drug-resistant bacteria are urgently required. Advanced nanomaterials are particularly promising due to their drug loading/releasing capabilities combined with their potential photodynamic/photothermal therapeutic properties. In this review, 0-dimensional, 1-dimensional, 2-dimensional, and 3-dimensional nanomaterial-based systems are comprehensively discussed for bacterial-based diagnostic and treatment applications. Since the use of these platforms as antibacterials is relatively new, this review will provide appropriate insight into their construction and applications. As such, we hope this review will inspire researchers to explore antibacterial-based nanomaterials with the aim of developing systems for clinical applications.

1. Introduction

The development of antibiotics in the 20th century resulted in the successful treatment of bacterial infections, which improved the quality of life of millions of patients worldwide.1–8 Unfortunately, their overuse and misuse has led to a rise in drug-resistant bacteria, which has rendered numerous antibiotics ineffective. This continuing trend poses a serious threat to the antibiotic development pipeline, which could eventually result in an inability to treat bacterial infections.9–14 If not appropriately addressed, there are significant concerns that we will return to a “pre-antibiotic” era.15,16 Therefore, new and effective treatment strategies are urgently required, which will result in a reduced risk of developing drug-resistant bacteria.

In the area of materials sciences, graphene and various other carbon-based nanomaterials have emerged as excellent components for conductor and battery-based applications.17–19 The significance of these findings has inspired the design of new and improved nanomaterials and the identification of valuable optical and thermal properties.20,21 These important characteristics have attracted considerable interest from biomedical researchers who wish to repurpose these nanomaterials for diagnostic and therapeutic applications.22–25 Nanomaterials have now been tailored towards a range of biology-based applications, including use as drug delivery systems (DDSs) in order to enhance the selectivity and efficacies of specific therapeutics.26–29 In addition, nanomaterials have exhibited the ability to perform photodynamic/photothermal therapy (upon light irradiation, PDT uses photosensitizers to convert environmental oxygen to reactive oxygen species (ROS), and PTT converts photoenergy to heat upon light activation, respectively) for disease treatment.30 Moreover, nanomaterials co-loaded with therapeutic agents have been shown to provide synergistic effects with PDT/PTT, which reduces the required therapeutic dose and minimalizes off-target toxicities,31 while theranostics (combined therapeutic and diagnostic) has been developed where imaging agents have been loaded onto nanomaterials to facilitate the ability “sense and treat”.

Nanomaterials are defined as materials with at least one length dimension being less than 100 nm.32 Recently, several reviews summarizing the development of nanomaterials for antibacterial applications have been reported. Niu and coworkers reviewed the antibacterial properties of different...
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metals and metal oxides including titanium derivatives, zinc oxide, nickel, copper and copper oxide, gold, palladium, selenium and iron.33 In another review, Truong and co-workers summarized the antibacterial mechanisms of different metallic nanomaterials.34 One of the most common mechanisms involves physical interaction with the bacterial surface, which kills bacteria through membrane damage. Which, results in ion release that causes enzyme inhibition or nucleic acid degradation, and ROS production. The authors concluded that there is an emerging trend for developing stimulus-activated nanomaterials, which require an external triggering element for the materials to function, affording “activatable” antibacterial materials. The most extensively used triggering elements include light (PDT/PTT) and magnetic force. Focusing on a variety of mechanisms facilitating antibacterial function, Song and co-workers prepared a comprehensive review on polymeric, antibiotic-free materials.35 The diverse antibacterial mechanisms include suppression of bacterial metabolism, catalytic bacterial killing by nanozymes and drug-loading/releasing. Moreover, the real-world applications of the as-developed strategies were deliberated for wound dressings, medical implants and food packaging. However, to the best of our knowledge, a summary of antibacterial nanomaterials in terms of their dimensionality has not been published (Scheme 1).

Dimensionality is an important asset of nanomaterials, which can change the intrinsic properties of the materials. Low dimensional materials including 0-D (zero-dimensional), 1-D (one-dimensional), 2-D (two-dimensional) and 3-D (three-dimensional) nanomaterials have attracted the interest of materials physicists and chemists owing to their exceptional mechanical, magnetic, electric and optical properties. Significantly, differences in dimensionality could lead to different biological activities for the low-dimensional materials. The low-dimensional materials and their antibacterial mechanisms covered in this review are described in Table 1.

The dimensional difference of the materials causes their antimicrobial mechanisms to differ. For example, 0-D nanomaterials can serve as both a metal ion-releasing therapeutic agent and carrier for antibiotic delivery. Due to their small size, 0-D nanoparticles can also be incorporated into hydrogels or loaded onto flat materials for practical applications.36 A representative clinical application is the use of a silver-nanoparticle-based hydrogel for treatment of bacterial infection on the skin.33,34 1-D nanomaterials are upgraded in respect to surface area and are free from the drawbacks of internal deposition compared to 0-D, while they share properties with both 0-D and their 2-D counterparts. 1-D rod-like nanomaterials are similar to 0-D with respect to performance and applications, while 1-D ribbon-like nanomaterials are more similar to 2-D because they are mostly derived from graphene.37 2-D nanomaterials owing to their large surface area have been used for the loading of antibiotics and other therapeutic agents and combined with PDT/PTT. They can also be used for the direct eradication of bacteria by physical cutting as assisted by their sharp edge.37 2-D materials are also degradable in human bodies thus avoiding the toxicity brought about by 0-D or 1-D nanomaterials.38 3-D nanomaterials are more likely to be a combination of 0-D, 1-D, or 2-D components, so they could exhibit the combined properties of 0-D, 1-D and 2-D nanomaterials resulting in composite systems with improved antibacterial properties.39 With the continuing development of materials science, the biocompatibilities of different nanomaterials have been greatly improved, which will stimulate the development of enhanced properties for improved clinical applicability. Within this review, to allow ease of understanding, we have separated antibacterial nanomaterials according to their dimensionality. In our review, we will discuss stimulus-activated and target-responsive low-dimensional nanomaterials that have been recently developed for antibacterial applications, with particular emphasis on the parameters that determine their practical applicability including the antibacterial spectrum, biocompatibility for clinical use and selectivity towards other pathogens.

2. 0-D nanomaterials

0-D nanomaterials are spherical-like materials with good dispersity and stability. These include metal-based nanoparticles,40,41 fullerenes, carbon dots, polymeric nanoparticles (polymer dots) and graphene quantum dots (GQDs).42,43 0-D nanomaterials are particularly attractive for antibacterial applications due to their high surface to volume ratios (drug loading/functionalisation) and ideal photophysical properties such as photoluminescence and PDT/PTT. 0-D nanomaterials originated from research on metal nanoparticles, which could dissociate metal ions and exhibit an inherent antibacterial effect. Representative 0-D material-based antibacterial agents are silver nanoparticles (AgNPs), which have inherent therapeutic properties towards bacterial infection with possible mechanisms being not only metal dissolution but also electrostatic adsorption and photocatalysis.44,45 AgNPs are one of the first examples of a non-chemotherapy-based approach used in both fundamental and clinical research.46,47 In recent years, researchers have focused on enhancing the therapeutic potential of AgNPs. This includes the use of nanocrystalline cellulose (NCC) to generate AgNPs/NCC systems that display superior antibacterial activity when compared to AgNPs alone.48 Huang and co-workers synthesised stable AgNPs using γ-irradiation in the presence of a chitosan solution, which acted as a “green” stabilizing agent. This method generated AgNPs with significant antioxidant

Scheme 1 Schematic illustration of the low-dimensional materials, including their antibacterial mechanisms, covered in this review.
Nanorods PEG-functionalised AuNRs (PU-Au-PEG) 

Inhibition of bacterial adhesion and PTT for multidrug-resistant bacteria (MDR)  

Nanofibers Cu²⁺-based coordination polymer nanofibers  

Disruption of membrane integrity inducing DNA condensation  

Nanotubes AgNO₃ mixed with N,N-bis(pyridyl-4-methyl)-N-fluorenyl-9-methoxycarbonyl [Fmoc]-3-glutamate (4MPFG) leading to gelation (4MPFG)  

Porphyridium-based porphyrin (Pp4N) mixed with AgNPs/NCC  

Table 1  Reported antibacterial mechanisms of low-dimensional materials covered in this review

| Composition of nanomaterials | Antibacterial mechanism | Ref. |
|-----------------------------|-------------------------|------|
| **0-D nanomaterials**       |                          |      |
| AgNP-based nanomaterials    |                        |      |
| Nanocrystalline cellulose (NCC)-coated AgNPs (AgNPs/NCC) | AgNP-triggered oxidative stress, protein dysfunction, and membrane and DNA damages | 48   |
| μ-Cysteine-functionalised AgNPs (μ-Cys-AgNPs) | μ-Cysteine inhibits biofilm formation, thus enhancing the therapeutic effect of AgNPs | 50   |
| Dextrin-based nanocomposite hydrogels (Dex-G3-Ag) | Controlled release of AgNPs, inducing oxidative stress, protein dysfunction, and membrane and DNA damages | 57   |
| Template-guided synthesis of ultrafine Ag NPs of around 2 nm using water-soluble and biocompatible γ-cyclodextrin metal–organic frameworks (CD-MOFs) | Ag⁺ ion release leading to protein dysfunction, and membrane and DNA damages | 54   |
| Integrating silver nanoparticles in situ into hydrogel materials (AgNP-hydrogel) | Kinetically-controlled release of antibacterial silver ions | 60   |
| **Nanomaterials based on other metallic and carbon-based particles** | Ag-Bi@SiO₂ significantly increased local temperature due to light absorption of BiNPs, leading to disruption of cell integrity and acceleration of silver ion release | 51   |
| Mesoporous silica-supported Ag-bismuth (Bi-Ag-Bi@SiO₂) | Inhibition of the function of extracellular ATP by CeO₂ nanoparticles, leading to disruption of initial bacterial adhesion. In addition, planktonic bacteria are killed by cytotoxic reactive oxygen species (ROS) generated by the MOFs | 70   |
| CeO₂-decorated nanoparticle MOFs (MOF@CeO₂NPs) | Programmable cell death of bacteria induced by carbon nanodots (C-dots) with different surface charges Electrostatic interaction between phosphate groups in the cell membrane of bacteria and the positive charges of the poly(ionic liquid) enhanced the cell membrane permeability, causing leakage of cytoplasmic contents. | 71   |
| Carbon nanodots (CNDs) (CND-250) | Dual PTT/PDT effect killing drug-resistant bacteria | 78   |
| **Nanomaterials based on polymeric particles** | Silver ion release by dissolution of the material after the oxidation of metallic silver | 86   |
| Poly(ionic liquid)/PVA hydrogel | Peroxidase-mediated transformation of low concentrations of H₂O₂ to toxic ROS species | 96   |
| Dual-mode antibacterial conjugated polymer nanoparticles (DMCPNs) | ROS generation and release of Cu²⁺ ions | 89   |
| Organic nanoparticles based on polymers that have a hydrophobic skeleton and hydrophilic side chain modified with protonated primary amines (PDCP) | Inhibition of bacterial adhesion and PTT for multidrug-resistant bacteria | 91   |
| Porphyridium-based porous organic polymer, FePPOPOH$_{20}$ | Dual-mode PTT/PDT | 92   |
| **1-D nanomaterials**       | Silver ion release by dissolution of the material after the oxidation of metallic silver | 86   |
| Nanowires  AgNWs | Peroxidase-mediated transformation of low concentrations of H₂O₂ to toxic ROS species | 96   |
| Co-V mixed metal oxide (MMO) nanowires (Co₃V₂O₈) | ROS generation and release of Cu²⁺ ions | 89   |
| **Nanofibers** Cu²⁺-based coordination polymer nanofibers (CuO/MnO₂ core nanostructures) | Inhibition of bacterial adhesion and PTT for multidrug-resistant bacteria | 91   |
| **Nanorods** PEG-functionalised AuNRs (PU-Au-PEG) | Dual-mode PTT/PDT | 92   |
| Bi₁₃S₉-coated AuNRs (Au@Bi₁₃S₉) | Firm bacterial adhesion causing cell wrapping and morphological disruption | 94   |
| γ-AlOOH, γ-MnOOH, and γ-Mn₃O₄ nanorods | Disruption of membrane integrity inducing DNA condensation | 87   |
| **Nanotubes** AgNO₃ mixed with N,N-bis(pyridyl-4-methyl)-N-fluorenyl-9-methoxycarbonyl [Fmoc]-3-glutamate (4MPFG) leading to gelation (4MPFG) | Dual-mode PTT/PDT | 103  |
| Nanoribbon-based supra-structure | Polycationic porphyrin (Pp4N) mixed with GNR-PEO2000 (Pp4N/GNR) | 103  |
| **2-D nanomaterials**       | GO destructs bacteria by cell membrane damage through chemical reactions, whereas rGO induces mechanical stress and pierces the cell membrane | 109  |
| Graphene-based nanomaterials | Bacterial structure damage and production of ROS, causing cytoplasm leakage and metabolic disorder | 117  |
| GO and reduced GO (rGO) | Membrane damage | 118  |
| Loading of sodium 1-naphthalenesulfonate (NA) onto reduced GO for chelation of AgNPs, producing AgNP-NA-rGO (AgNP-NA-rGO) | Physical disruption of bacterial membrane | 112  |
| PEG-functionalised GO mixed with AgNPs (GO-PEG-Ag) | Membrane damage, and oxidation of intracellular proteins and lipids | 136  |
| Loading of GO with AgNPs and CoFe₂O₄NPs (Ag-CoFe₂O₄-GO) | PDPT | 128  |
| GO nanocomposites assembled with QASs (quaternary ammonium salts) | Membrane adhesion followed by photothermal antibiotic release | 123  |
| **2-D MoS₂-based nanomaterials** | AgBrNPs mixed with MoS₂ nanosheets AgBr@MoS₂ | 109   |
| Chitosan-coated 2-D MoS₂ (CS@MoS₂) | Quaternized chitosan (QCS)-modified MoS₂ nanoflakes (QCS-MoS₂) | 123   |
activity against Gram-positive and Gram-negative bacteria.\textsuperscript{49} Motivated to develop a more clinically relevant system, Ju and co-workers developed \( \nu \)-cysteine-functionalised AgNPs (\( \nu \)-Cys-AgNPs) for the enhanced treatment of planktonic bacterial biofilms (Gram-positive and Gram-negative). Biofilms are complex microbial communities encapsulated by extracellular polymeric substances (EPSs), which support bacterial growth and provide protection against antibacterial treatments. Using this strategy, \( \nu \)-Cysteine (\( \nu \)-Cys) inhibited biofilm formation, resulting in the dispersion of bacteria, which enhanced the therapeutic effect of the AgNPs. This approach was called the “disperse-then-kill” strategy.\textsuperscript{50} \( \nu \)-Cys-AgNPs displayed significant therapeutic efficacy towards \textit{E. coli}, \textit{S. aureus}, and \textit{P. aeruginosa}, including their hard to treat biofilms. In addition, Ag-based 0-D nanomaterials have been used in combination with other systems to improve the overall therapeutic efficacy and to reduce side effects. One particular example includes the development of mesoporous silica supported Ag–bismuth (Bi) (Ag–Bi@SiO\(_2\)) nanoparticles by Dong and co-workers.\textsuperscript{51} The light irradiation (808 nm, 15 min, and 1 W cm\(^{-2}\)) of Ag–Bi@SiO\(_2\) resulted in a significant temperature increase (PTT) due to the light absorption of the BiNPs. This temperature increase accelerated the release of Ag\(^+\) ions from the silica shell and provided an enhanced antibacterial effect. Using this approach, methicillin-resistant \textit{S. aureus} (MRSA) and its biofilms were eradicated. Importantly, the clinical significance of Ag–Bi@SiO\(_2\) NPs was successfully demonstrated using mouse skin-infection models (Fig. 1). Other strategies include the combination of AgNPs with magnetic iron oxide Fe\(_3\)O\(_4\)/Fe\(_2\)O\(_3\).\textsuperscript{52,53} Due to the challenges involved in synthesising and controlling the size and colloidal stability of AgNPs, Zhang and co-workers developed a template guided approach for the synthesis of ultrafine AgNPs using water soluble \( \gamma \)-cyclodextrin (\( \gamma \)-CD) metal–organic frameworks (CD-MOFs).\textsuperscript{54} Peptide functionalisation (GRGDs (Gly-Arg-Gly-Asp-Ser)) of the MOF-based system facilitated platelet adhesion and aggregation at the injury site through peptide binding to the integrin GPIIb–IIIa receptor. This reduced the time for blood to clot and
Hydrogels are attractive for effective wound management, due to their excellent biocompatibility and therapeutic loading ability, including the incorporation of nanoparticles.\textsuperscript{55–58} More specifically, stimulus-responsive/smart hydrogels are particularly attractive for a range of biomedical applications;\textsuperscript{59} however, long-term stability issues are often observed. To overcome this, Zhao and co-workers demonstrated that the simple loading of AgNPs into colour-responsive hydrogels prevented bacterial adhesion, hydrogel damage, and maintained the colour-responsive properties during application and long term storage.\textsuperscript{60} More recently, Dai and co-workers reported on the design of an acid-responsive hydrogel, Dex-G5-Ag, for the controlled release of AgNPs for the treatment of bacterial infection.\textsuperscript{57} Dex-G5-Ag hydrogels were formed by Schiff-base condensation between dextran (Dex-CHO) and an amino-functionalized dendrimer, G5-Ag (AgNPs encapsulated). The as-formed hydrogel was shown to degrade in an acidic environment and release AgNPs (Fig. 2). This strategy exhibited minimal cytotoxicity towards normal cell lines and proved effective for the treatment of a variety of bacteria including \textit{E. coli}, \textit{P. aeruginosa}, \textit{S. aureus}, and \textit{S. epidermidis}, and the effects were further evaluated using mouse-infection models. Other examples include the use of poly(ionic liquid)/poly(vinyl alcohol) (PVA) hydrogels for the development of antibacterial wound dressings.\textsuperscript{58}

However, silver as a first-generation antibacterial material exhibits clear toxicity towards tissues, since it can be deposited in bodies leading to heavy-metal accumulation, which makes it hard for the approach to be clinically-accepted. Effective as it is, it could also kill other cells due to low selectivity. Functionalization or encapsulation of silver could to some extent solve the problem but this approach is still far from satisfactory. Over the last several years, researchers have explored and evaluated the ability of non-silver-based nanomaterials for the treatment of bacteria. Firstly, other metals were evaluated, which mainly included a similar but much milder metal: gold. Gold nanoparticles (AuNPs) are well-established multifunctional 0-D nanomaterials used for biosensing, as drug carriers, as well as for theranostic, PDT/PTT and immunology applications.\textsuperscript{61–66} Perry and co-workers reported a unique antibacterial AuNP strategy, in which gold ions (\textit{AuCl}_4^-) were reduced and capped by cefaclor a second-generation antibiotic (Fig. 3).\textsuperscript{65} AuNP-cefaclor was shown to have potent antibacterial activity towards \textit{S. aureus} and \textit{E. coli}, while the individual use of AuNPs and cefaclor had minimal therapeutic effects (Fig. 3). Similarly, Jiang and co-workers explored AuNPs modified with 4,6-diamino-2-pyrimidinethiol (DAPT) as antibacterial agents (DGNPs).\textsuperscript{67} More recently, various particle sizes of DGNPs were evaluated against bacteria in order to correlate particle size with antibacterial activity.\textsuperscript{67} The antibacterial effects were assessed against Gram-negative and Gram-positive bacteria. Ultrasmall DGNPs (< 2 nm (uDGNPs)) displayed broad spectrum activity and as a result they were incorporated into agarose gels for wound dressing applications. The same authors evaluated a range of N-heterocyclic molecules with AuNPs, which displayed broad spectrum activity. Optimised antibacterial activities were achieved by varying the molar ratios of N-heterocycles with the precursor of AuNPs, HAuCl\textsubscript{4}.\textsuperscript{68} Compared to silver, gold-based agents are particularly delicate towards tissues, but this is also accompanied with a weakened antibacterial effect. Consequently, effective gold-based approaches are always assisted by using co-loaded antibiotics or other agents in order to attain acceptable performance.

Apart from the issues discussed above regarding AgNPs and AuNPs, these 0-D nanomaterials are expensive and result in metal ion release into the environment. These limitations
organisms. More specifically, ATP plays a crucial role in bacterial
affinity of the lanthanide cerium (Ce) for ATP. CeO
2 prevented
adhesion and biofilm formation. Exploiting the known binding
triphosphate (ATP) is an essential small molecule for all living
light irradiation of the MOF@CeO
2 NPs facilitated the production
biofilm formation through ATP extraction. Subsequent 638 nm
and image bacteria.71 These antibacterial CNDs were developed
example was demonstrated by Yang and co-workers, in which
was facilitated through a MOF consisting of zirconium (Zr) clusters
combination therapy using the MOF@CeO
2 NPs was shown to be an
effective treatment for bacteria and biofilm inhibition.

In order to further remove the toxicity of metals, other
inorganic material systems were developed. A representative
example was demonstrated by Yang and co-workers, in which
carbon nanodots (CNDs) were designed to simultaneously treat
and image bacteria.71 These antibacterial CNDs were developed
using the wide-spectrum antibiotic metronidazole72 as the sole
carbon source. CNDs-250 were prepared at 250 °C for 3 h and
8 h, respectively (Fig. 4a). Evaluation of the optical density (OD)
of bacterial solutions demonstrated a 71.7% inhibition of
P. gingivalis at low concentrations (1.25 µg mL
−1), similar to
that of metronidazole (Fig. 4b). In contrast, when using control
systems or treating other bacteria (Fig. 4c–e), the inhibition
effects disappeared, thus demonstrating a selective effect
P. gingivalis only. Based on this observation, the effects
of CNDs-250 were further screened against a variety of bacteria,
and it was found that CNDs-250 displayed significant activity
towards obligate anaerobes (P. micros, P. intermedia, P. gingivalis
and Fusobacterium) rather than facultative anaerobes (S. mutans),
obligate aerobes, or microaerophilic (E. coli) bacteria, which is
similar to metronidazole which is the carbon source of the
CNDs-250. The low toxicity towards human cell lines combined
with their fluorescence properties enabled their use in cell
labelling and imaging experiments. Importantly, CNDs were
shown to be selective for bacteria in comparison to metronida-
only, displaying 60% cell viability in MC3T3-E1 cells. This
underscores the significance of nanomaterials for antibacterial
applications. MC3T3-E1 cells incubated with CNDs-250
displayed clear fluorescence emission when excited using green,
blue, and UV light excitation, respectively. Another example
reported by Qu and co-workers,73 evaluated the programmed
cell death of bacteria induced by carbon dots (C-dots) with
different surface charges. This report provides unique insights
into the design of C-dots for antibacterial applications.

Yang and co-workers found that GQDs derived from graphene
oxide (GO-GQDs) displayed weak antibacterial activity. This was
believed to be due to their zero Gaussian curvature. In their
study, they rationalised that the preparation of GQDs from C60
would provide a non-zero Gaussian curvature and provide
improved antibacterial activity. These C60-GQDs exhibited
significant antibacterial activity against S. aureus only
(Fig. 5a),74 which was believed to be a result of the disruption
of the bacterial cell envelope (confirmed by TEM images). Interestingly,
E. coli was shown to have smooth surfaces
regardless of the type of GQDs used (Fig. 5b). Scanning electron
microscopy (SEM) images indicated that C60-GQD-treated
S. aureus cells were coated with NPs, whereas, GO-GQD-treated
S. aureus cells and all E. coli cells (no matter which type of GQDs
was used to treat them) remained smooth (Fig. 5c). This select-
ivity for S. aureus demonstrated the importance of both the
nanomaterial source and shape for physical contact-mediated
treatment of bacterial species (i.e. S. aureus).

However, since all metallic and non-metallic elements are
inevitably incompatible with tissues and can to some extent
stay inside bodies for a long time, people have started to
explore the use of polymers. Polymeric nanoparticles provide
a low cost, highly degradable, and biocompatible alternative.
Moreover, the homogeneity of polymeric nanoparticles is much
better than metal nanoparticles, thus ensuring the stability of
the antibacterial activity. A recent example was reported by Qin
and co-workers,75 in which guanidine-based nanogels were
explored using an infected mouse model. In addition, these
nanogels were applied to the design of antimicrobial materials.
A similar polymeric nanoparticle strategy was reported, which
employed the antimicrobial functionality of N-halamine.

![Fig. 4](https://example.com/fig4.jpg)

**Fig. 4** (a) Schematic illustration of CNDs-3h and CNDs-8h prepared from
metronidazole and differing by reaction time. (b and c) Growth curves of
*P. gingivalis* after incubation with various concentrations of metronidazole
and CNDs-250 (b), and CA-CDs (c) for 24 h. (d and e) Growth curves of
*S. mutans* (d) and *E. coli* (e) after incubation with various concentrations
of CNDs-250 for 24 h. Images reprinted with permission from ref. 71. Copyright 2017, The Royal Society of Chemistry.
propoxy]benzaldehyde (BFPB) that exhibited peroxidase-like activity toward a peroxidase substrate 3,3',5,5'-tetramethylbenzidine (TMB) with H$_2$O$_2$ under 808 nm NIR irradiation. While Cheng and co-workers in 2021 developed a polymer–antibiotic conjugate consisting of penicillin, 2-chloroethyl methacrylate (CEMA), and hydroxethyl methacrylate (HEMA) crosslinked together and self-assembled into particles, as a strategy to develop resin-based restorative dental materials for sustained antibacterial therapy. $^{84}$ S. mutans biofilms resulting in dental caries were cultured and eradicated using the as-developed polymers, the slow antibiotic release discouraging bacterial resistance and ensuring long-term efficacy in the prevention of dental caries. Upon exposure to an enzymatic challenge resembling true intra-oral conditions, the efficiency of the antibacterial agent was maintained, demonstrating the potential for long-term effects and effective clinical behaviour.

### 3. 1-D nanomaterials

1-D Nanomaterials are linear-based materials, which could have both stability/dispersity and capacity to load functional agents. Compared to 0-D, 1-D nanomaterials have a greatly-enhanced surface area and inherent photo-triggered therapeutic effects. These advantages have aroused significant interest and resultant development of these materials. Examples of such nanomaterials include nanotubes, nanowires, nanoribbons, nanofibers and nanorods, all of which exhibit exceptional thermal, electronic, mechanical, optical and magnetic properties. However, in comparison to 0-D nanomaterials, the development of 1-D nanomaterial technology was initially slow due to difficulties in synthesis and control of their morphology. $^{84}$ 1-D nanomaterials are now fully established and used routinely in materials sciences and for biomedical applications. $^{83,84}$ Firstly, ball-shaped metal particulates have been defined as 0-D, while rod-like materials are the archetypical 1-D nanomaterials, which initiated the 1-D materials area. Given that the main difference between these 0-D and 1-D metals is shape, metal-based 1-D rod-like metals are similar to 0-D metal nanoparticles in many aspects, and are attractive platforms for the development of effective antibacterial agents, $^{85}$ which is a result of their inherent antibacterial activity (i.e. Ag and Cu ions). $^{86}$ Recent reports include the electron beam (e-beam) irradiation of silver nanowire (AgNW) films. Surface irradiation (1200 kGy) altered the AgNW film surface morphology and chemical composition, which enhanced its antibacterial activity towards both E. coli and S. aureus. $^{86}$ This example represents a simple method for enhancing the antibacterial properties of nanomaterials, however, the inherent toxicity of silver is still a big concern. In order to reduce toxicity, the approach is to control the release of silver. Liu and co-workers reported antibacterial Ag(i) metallogels formed by the self-assembly of nanotubes and nanofibers, confirmed by SEM and TEM images. The addition of an aqueous solution of AgNO$_3$ to N,N-bis[pyridyl-4-methyl]-N-fluorenyl-9-methoxycarbonyl (Fmoc)-L-glutamate (4MPFG) resulted in instant gelation at room temperature. These metallogels displayed antibacterial activity...
against Gram-positive and Gram-negative bacteria and toxicity was reduced due to gel incorporation.\(^{87}\) Interestingly, this self-assembly was largely dependent on the gelator concentration, resulting in the formation of nanostructures ranging from nanotubes to nanofibers. Interestingly, the self-assembly of nanofibers was shown to result in increased damage to bacterial cell membranes when compared with nanotubes, which correlated with enhanced antibacterial activity. This research demonstrates the importance of nanomaterial morphology for efficient antibacterial applications.

With the aim of enhancing biocompatibility towards tissues, copper (Cu)-based and Au-based 1-D nanomaterials were proposed. Copper (Cu)-based 1-D nanomaterials include Cu/C, CuO nanorods and nanoplatelets, which have exhibited comparable antibacterial efficacies to known antibiotics with enhanced biocompatibility.\(^{88}\) More recently, Rauf and co-workers reported Cu\(^{2+}\)-based coordination polymer nanofibers,\(^{89}\) that were formed from \([\text{Cu(HBTC)}(\text{H}_2\text{O})_3]\)\(^{2+}\) and HBTC (1,3,5-benzenetricarboxylic acid) (Fig. 6a). The nanofibers exhibited excellent antibacterial effects against both \(E.\) coli and \(S.\) aureus (Fig. 6b). The proposed antibacterial mechanism was the generation of ROS combined with the release of Cu\(^{2+}\) ions. As with AuNPs, gold nanorods (AuNRs) have been shown to exhibit a range of diagnostic and therapeutic applications.\(^{90}\) A recent study by Zhao and co-workers demonstrated PEG-functionalised AuNRs for the PTT treatment of multidrug-resistant \(S.\) aureus and \(P.\) aeruginosa\(^{91}\) biofilms. These functionalised AuNRs were used with infected mouse models in vivo. Similarly, Bi\(_2\)S\(_3\)-coated AuNRs (Au@Bi\(_2\)S\(_3\)) have been reported by Wang et al. for the light-based treatment of bacteria.\(^{92}\) The NIR light irradiation (808 nm) of Au@Bi\(_2\)S\(_3\) resulted in the PDT/PTT-based treatment of \(E.\) coli and \(S.\) aureus (Fig. 7a), with clear inhibition seen in bacterial cultures (Fig. 7b). Interestingly, the individual use of each component resulted in minimal antibacterial activity being observed. As such, each component was required to have a good therapeutic effect.

In the area of 1-D research, oxides of metals have been investigated. For example Mn\(_3\)O\(_4\)-based nanorods and nanotubes have been developed by Chen et al., through a bi-directional-bi-dimensional growth model with biocompatibility comparable to gold systems.\(^{93}\) The antibacterial properties were confirmed using a range of bacteria including \(B.\) subtilis, \(S.\) aureus, \(S.\) faecalis, \(P.\) aeruginosa, and \(E.\) cloacae. The research further illustrated the importance of the morphology of nanomaterials, since Mn\(_3\)O\(_4\) nanotubes were found to have a greater bacterial inhibition towards Gram-negative bacteria, than Mn\(_3\)O\(_4\) nanorods. With a growing interest in 1-D nanomaterials as bacterial-resistant materials and for antibacterial applications, Selim and co-workers developed synthetic methods for the preparation of \(\gamma\)-AlOOH, \(\gamma\)-MnOOH, and \(\alpha\)-Mn\(_2\)O\(_3\) nanorods exhibiting biocompatibility/stability comparable to those of Mn\(_3\)O\(_4\) nanorods.\(^{94}\) The antibacterial activity was evaluated against \(P.\) aeruginosa, \(S.\) aureus, \(E.\) coli, \(B.\) subtilis, \(B.\) pertussis, and \(C.\) albicans, and all three nanorods exhibited significant antibiofilm activity, and \(\alpha\)-Mn\(_3\)O\(_4\) was particularly effective against \(B.\) subtilis, \(P.\) aeruginosa, and \(B.\) pertussis biofilms. In particular the authors highlighted the importance of controlling the shape and the surface area of nanomaterials for achieving appropriate

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**Fig. 6** (a) Basic schematic of the growth of the \([\text{Cu(HBTC)}(\text{H}_2\text{O})_3]\)-based nanofibers. (b) Images of \(E.\) coli and \(S.\) aureus treated with different concentrations of \([\text{Cu(HBTC)}(\text{H}_2\text{O})_3]\)-based nanofibers. Images reprinted with permission from ref. 89. Copyright 2019, The Royal Society of Chemistry.

**Fig. 7** (a) Construction of Bi\(_2\)S\(_3\)-coated AuGNRs for the PDT/PTT-based treatment of bacteria. (b) Images of \(E.\) coli and \(S.\) aureus colonies on nutrition cultures after treatment with Au@Bi\(_2\)S\(_3\) with and without laser irradiation. Images reprinted with permission from ref. 92. Copyright 2020, Elsevier B.V.
antibacterial efficacy. It is important to note, that many researchers have endeavoured to improve the antibacterial efficacy of 1-D nanomaterials through functionalisation with antibacterial peptides.95 Wang and co-workers reported Co-V mixed metal oxide (MMO) nanowires, which consisted of Co3V2O9 dispersed amongst Co3O4.96 Exploiting the intrinsic oxidase-like and peroxidase-like catalytic activities of Co-V MMO nanowires, in combination with low concentrations of H2O2 (50 μM), resulted in the successful treatment of E. coli. The antibacterial activity was attributed to the peroxidase-mediated transformation of H2O2 to the more harmful superoxide O2−. These materials offer an alternative to expensive Ag- and Au-based nanomaterials; however the homogeneity of suspension requires additional improvement. Importantly, the ability to use low concentrations of H2O2 minimises healthy tissue damage.

To solve the chronic problems caused by metals, 1-D researchers have developed systems from inorganic materials. The incorporation of non-metal/metal components into 1-D nanomaterials results in improved properties for sensing and therapeutic applications (e.g. electrochemical, fluorescence and PTT).97 Within this realm, graphene-derived 1-D nanomaterials have shown promise for antibacterial applications with not only PTT or agent-loading functions but also, a physical-cutting mechanism to assist therapy.98,99 For example, Davis and co-workers reported the combination of single-walled nanotubes (SWNTs) and lysozyme (displaying inherent antibacterial activity for Gram-positive bacteria) for the construction of multicomponent fibers.83 The cationic surfactant, tetracyclohexylmethyloxonium bromide was used to improve the stability of dispersions and enhance mechanical properties. CNTs are particularly attractive due to their tuneable thermal and electrical characteristics and high surface area to volume ratio. Additionally, CNTs have been shown as effective carriers for metallic NPs as well as increasing their aqueous stability and therapeutic properties. Hidal and co-workers reported the coating of multi-walled carbon nanotubes (MWCNTs) with AgNPs to develop an effective tool to prevent membrane fouling. In their study, the incorporation of AgNP-CNTs into polyethersulfone (PES) ultrafiltration (UF) membranes was efficient for antifouling applications with significant inhibitory activity towards both E. coli and S. Aureus being observed.100 With the AgNPs co-loaded into CNTs, the inherent toxicity was also avoided and the selectivity was improved. Hao et al. reported MWCNT–glucosamine–AgNP nanocomposites for the construction of functional materials with long-term antibacterial activity. Using this strategy, MWCNTs were grafted with glucosamine, which facilitated the coordination of AgNPs (Fig. 8a–d).101 This approach permitted the slow and controlled release of AgNPs, which provided long-term antibacterial activity against S. aureus over 35 days (Fig. 8e) with minimal side effects.

When compared with antibacterial drugs, the effects of 1-D nanomaterials that rely solely on photophysical mechanisms are insufficient. Within our group, graphene nano-ribbons (GNRs) have been evaluated for PDT/PTT applications. GNRs exhibit excellent mechanical strength and good biocompatibility and with co-loaded PDT agent can afford potent antibacterial effects comparable to those of antibacterial drugs.102,103 Through our work, we have developed a series of structurally well-defined GNRs functionalised with hydrophilic flexible poly(ethylene oxide) (PEO) chains to reduce the toxicity and improve hydrophilicity.85 GNR-PEO exhibited aqueous dispersion/stability suitable for biological applications. Evaluation of the morphology of these systems revealed hierarchical self-assembled GNR-PEO aggregates as supramolecular nanostrips, which subsequently formed ultralong nanobelts. In addition, the near-infrared (NIR) absorption (750–850 nm) of GNR-PEO permitted its use as PTT agents. PTT was demonstrated in combination with a porphyrin (PDT agent) as a dual wavelength (660 + 808 nm) PDT/PTT therapeutic.103 Specifically, polycationic porphyrin (Pp4N) was mixed in aqueous solution with GNR-PEO2000, which resulted in π–π stacking interactions and formation of a Pp4N/GNR nanocomposite. The positively charged ammonium groups on Pp4N acted as targeting moieties for the negatively charged bacterial surfaces (Fig. 9a). Pp4N/GNR nanocomposite exhibited synergistic antibacterial effects under dual irradiation (660 nm + 808 nm) and was successfully applied for the treatment of A. baumannii and methicillin-resistant S. aureus (MRSA) infected wounds in mouse models (Fig. 9b).

4. 2-D nanomaterials

While 1-D nanomaterials can be used for numerous applications, many issues remain including limited surface area and loading capacity. Moreover, a lone PTT function could not generate significant clinical activity, which prevented them from becoming comprehensive therapeutics. Consequently, 2-D nanomaterials were developed as novel large-surface area and platform-like nanosystems with combined drug/PDT/PTT functionality. 2-D nanomaterials represent a class of nanomaterials that possess sheet/layered-like structures. Compared with 0-D and 1-D, these unique structural features afford unprecedented physical, electronic and chemical properties, which have led to their exploration in electronics/optoelectronics, electrocatalysis, batteries, supercapacitors, solar cells, photocatalysis, and sensing applications.104 Moreover, the sheet-like nature of 2-D platforms equips them with a large surface area, which is...
highly photoreactive with improved physical contact and PDT/PTT performance. As such these 2-D materials can be used to develop dressings, and can be co-loaded with 0-D, 1-D, drugs, or photosensitizing agents to provide synergistic effects. In general, due to the increased surface area, 2-D nanomaterials possess all the advantages of 1-D nanomaterials and in addition exhibit high agent-loading and are degradable/biocompatible when compared with 0-D nanomaterials. More specifically, a range of 2-D nanomaterials have been developed and explored for antibacterial applications. These include graphene-based, transition metal dichalcogenide, transition metal oxide and transition metal hydroxide 2-D nanomaterials. The large surface area of 2-D nanomaterials facilitates the incorporation of a range of therapeutics; therefore, in combination with their inherent electrochemical/fluorescent properties this enables the development of theranostics. In particular, the 2-D nanomaterial, graphene oxide (GO) has been extensively shown to have inherent antibacterial activity, by inducing ROS production (oxidative stress) and physical contact with bacteria resulting in damage to their cell membranes. A recent report by Chakraborty and co-workers has explored the physical cutting and oxidative stress mechanisms of both GO and reduced GO (rGO). The extent of bactericidal efficacy varying between Gram-positive and Gram-negative bacteria was found to be size-, shape-, and type-dependent. Gram-positive S. aureus was found to be more vulnerable to GO compared to Gram-negative P. aeruginosa. Within GO, the oxygen groups enable the wrapping of bacterial cells and inhibits the entry of nutrients. In addition, oxidative stress is generated on the cell membrane for destabilization and disintegration, which leads to leakage of cytoplasmic fluid. A recent study illustrated the significance of GO nanosheets for the treatment of MDR bacteria (“superbugs”). The MDR bacteria strains included E. coli, K. pneumoniae, S. aureus, P. aeruginosa, P. mirabilis, and S. marcescens. Due to the clinical importance of biofilms, He et al. evaluated the efficacy of GO towards S. mutans biofilms, in which concentration-dependent inhibition was observed for biofilm formation. However, no antibiofilm effect was observed for mature biofilms. This illustrates the need for further optimisation of graphene-based nanomaterials. Consequently, GO platforms were co-loaded with active small molecules and 0-D nanoparticles in order to enhance the antibacterial efficiency via synergistic effects. Motivated to enhance ROS production by GO, Zhou and co-workers loaded the phototherapeutic sodium anthraquinone-2-sulfonate (AQS) onto GO using π-π interactions to afford the nanocomposite AQS-GO (Fig. 10a). AQS-GO exhibited light-based inhibitory activity against E. coli, where clear cell damage could be seen when compared with GO under irradiation (Fig. 10b). Quaternary ammonium salts (QASs) are well-known antibacterial agents, as such, Ye and co-workers developed a GO nanocomposite using dodecyl dimethyl benzyl ammonium chloride. The QAS (positively charged) was used as a bacteria-targeting unit and an antibacterial agent. The system provided long-term antibacterial activity against both E. coli and S. aureus. Other reported strategies aimed at enhancing the therapeutic efficacy of GO include the loading of sodium 1-naphthalenesulfonate onto rGO for chelation of AgNPs, which afforded antibacterial AgNP–rGO hybrids. Remarkably, the strategy was superior to AgNP–NA–rGO. Other reported strategies aimed at enhancing the therapeutic efficacy of GO include the loading of sodium 1-naphthalenesulfonate onto rGO for chelation of AgNPs, which afforded antibacterial AgNP–rGO hybrids. The authors proposed that these AgNP–NA–rGO hybrids could be used in

Fig. 9 (a) Basic schematic of the Pp4N/GNR nanocomposite strategy for the light-based treatment (PTT/PDT) of bacteria. (b) Photographs of A. baumannii- and methicillin-resistant S. aureus (MRSA) infected wounds with and without treatment over a 12-day period. Images reprinted with permission from ref. 103. Copyright 2019, Wiley-VCH.

Fig. 10 (a) Schematic illustration of the preparation procedure of a composite antibacterial agent AQS-GO. (b) SEM images of E. coli incubated with GO and AQS-GO at 0 min and 160 min under dark (D) and visible light irradiation (L). Images reprinted with permission from ref. 112. Copyright 2019, Elsevier B.V.
sprayable antibacterial solutions. More recently, Hao and co-workers reported PEG-functionalised GO for the development of stable aqueous AgNP nanocomposites. Non-PEGylated GO with Ag was shown to irreversibly aggregate in aqueous solution, while AgNPs–GO–PEG displayed good aqueous stability, low cytotoxicity and long-term antibacterial activity (~95%) against E. coli and S. aureus (stored in saline solution for one week). Similarly, Zhou and co-workers reported the loading of GO with AgNPs and CoFe2O4NPs for both Pb2+ removal and bacterial absorption/eradication in contaminated water.

The combination of GO with other nanoplatforms results in systems with therapeutic and diagnostic (visualisation) capability. For example, Kim and co-workers have developed a GO-MoS2 nanocomposite film through stacking of 2-D MoS2 (molybdenum disulfide) onto GO. This system exhibited a time-dependent therapeutic effect against E. coli, which was visualised using holotomographic (HT) microscopy (Fig. 11). Alternatives to graphene-based materials include graphitic carbon nitride nanosheets, which exhibit antibacterial activities against E. coli, S. typhimurium, S. enteritidis, S. aureus, L. monocytogenes, B. subtilis, and B. cereus. The use of such graphitic carbon nitride nanosheets is still at the very early stage of development and as such considerably more research is required to evaluate these systems. In summary, the importance of graphene derivatives for biomedical research is attributed to excellent photothermal conversion efficiency, the capacity to co-load active agents, and the satisfactory biocompatibility. However, graphene systems are difficult to completely disperse in aqueous solutions without modification, meaning that other graphene-like 2-D materials are now being considered for development.

After the emergence of GO, a range of other 2D nanomaterials with properties similar to graphene/GO were developed, which are collectively known as graphene-like platforms and exhibit additional functionality. 2-D MoS2 is a popular graphene-like nanomaterial that has the advantages of graphene and exhibits improved stability/dispersivity. 2-D MoS2 exhibits good structural, physicochemical, optical properties and excellent biocompatibility. More importantly, MoS2 can be easily metabolized and decomposed in human bodies and is excreted rapidly, which is a significant improvement when compared to graphene derivatives when considering the body damage or side effects induced by these materials. Recently, Niu et al. reported a Gram-selective antibacterial 2-D MoS2 nanomaterial, which achieved selectivity by controlling the light-irradiation time (photomodulation). This is of particular significance since providing appropriate treatment for the right bacteria reduces the development of drug-resistant bacteria. As with GO, synergistic studies for the loading of small drug molecules and nanoparticles onto 2-D MoS2 for constructing composites have been performed. A recent report by Ji and co-workers focused on the combination therapy between antibiotics and PTT. In this study, ofloxacin (OFLX)-loaded 2-D MoS2 nanoflakes were used as a PTT/antibacterial platform. 2-D MoS2 nanoflakes were modified with electropositive quaternized chitosan (QCS) to improve the aqueous dispersion and act as a targeting moiety for the negatively charged surfaces of bacteria. OFLX-loading on 2-D MoS2 nanoflakes was achieved using π–π stacking and hydrophobic interactions. Remarkably, QCS–MoS2–OFLX proved effective as an antibiotic/PTT agent at low temperatures (45 °C) and low antibiotic concentrations. The effectiveness of the 2-D nanomaterial was demonstrated in vitro and in vivo for the treatment of MRSA (Fig. 12). Nitric oxide (NO) is a known signalling molecule found in the human body with inherent antibacterial properties. These properties are...
achieved through the known transformation of NO to more harmful reactive nitrogen species (RNS) i.e., peroxynitrite (ONOO−). Unlike traditional antibiotics, the antibacterial properties of NO are not dependent on the type of bacteria and in addition it is known to promote wound healing. Gu and co-workers rationalised that the use of a 2-D nanomaterial would facilitate the effective delivery of NO (nanovehicle) and improve the therapeutic effects against bacteria. As a result, α-CD modified 2-D MoS2 nanosheets were assembled with the heat sensitive (NO) donor N,N′-diethyl-4-phenylendiamine (BNN6). NIR light irradiation (808 nm) activated PTT and generated NO. The antibacterial effects were successfully demonstrated using infected mouse models.

2D MoS2 has been used for loading nanoparticles, in combination with AgNPs and β-Cys for the construction of an effective antibacterial nanocomposite. More recently, a multifunctional nanomaterial was developed consisting of 2-D MoS2 nanosheets and AgBr nanoparticles (AgBr@MoS2) grown on Ti-based implant materials; the AgBr was co-loaded to reduce the inherent toxicity. Visible light irradiation (660 nm) of the nanomaterial resulted in significant photothermal efficacy against (660 nm) of the nanomaterial resulted in significant photothermal efficacy against E. coli and S. aureus. 2-D Ti3C2T X MXene nanosheets have emerged as an attractive nanomaterial for biomedical applications. In 2016, Ti3C2T x MXene nanosheets were evaluated for their antibacterial properties and compared with GO. Significantly better antibacterial effects were observed when compared to those of GO (~2-4 fold greater) against both E. coli and B. subtilis. As with graphene, SEM and TEM images revealed physical damage to the cell membrane and biological assays identified the induction of oxidative stress. The authors anticipate that MXenes will find use in biofouling and bactericidal applications due to the good biocompatibility. Recently, Zhang and co-workers used the known low-cost and biocompatible semi-conductor material, Sb2Se3 for the development of an antibacterial nanomaterial. In this report, polyvinylpyrrolidone (PVP)-capped Sb2Se3 was used for the treatment of bacteria including E. coli and methicillin-resistant S. aureus (MRSA) in vivo. 2-D tungsten disulfide (WS2) nanomaterials have been explored for water filtration applications, exhibiting excellent antibacterial properties against S. aureus and E. coli. More recently, Pramanik and co-workers developed a combined 2-D and 0-D nanomaterial strategy for the identification of antibiotic-resistant bacteria. This was achieved through the construction of WS2-AuNPs and using the surface enhanced Raman spectroscopic properties of AuNPs enabling the rapid detection (90 min) of MDR Salmonella DT104. Ikram and co-workers have reported Zr-doped MoS2 nanosheets for catalytic and antibacterial applications. These systems were effective against E. coli and S. aureus through the catalytic generation of ROS. This catalytic-based antibacterial approach was then further elucidated by Yin and co-workers by directly loading enzymes onto 2-D MoS2. They reported lysozyme-coated MoS2 nanosheets (Ly-MoS2), which displayed good antibacterial efficacy against Gram-negative E. coli and Gram-positive B. subtilis. This was due to synergistic effects between the lysozyme and peroxidase activity of the MoS2 nanosheets.

Often medical implants are prone to bacterial infection; for this reason, Wu and co-workers developed a chitosan@MoS2 nanocomposite to overcome this problem. Chitosan@MoS2 was deposited onto medical Ti-based implants through an electrophoretic deposition method. Upon dual-light irradiation of chitosan@MoS2-TiC02 [660 nm and 808 nm], significant bacterial inhibition was observed in vitro and in vivo. Moreover, 2-D MoS2 could be combined with graphene derivatives. PEG-2-D MoS2/rGO therapeutic nanoflakes were developed by Li and co-workers and loaded with a streptomycin sulfate (SS) antibiotic. In combination with NIR light irradiation, the therapeutic efficacy of PEG-MoS2/rGO-SS was significantly enhanced and a synergistic therapeutic effect was observed between SS and light-based therapy. Black phosphorus (BP) is an emerging 2-D nanomaterial used for semiconductor applications. Aksoy and co-workers reported BP/AuNP nanocomposites for antibacterial applications with good biocompatibility. The NIR laser irradiation (808 NM) of these nanocomposites resulted in the PTT/PDT treatment of planktonic bacteria and biofilm-based E. faecalis (Fig. 13a). Live/dead staining and fluorescence imaging were used to evaluate the therapeutic efficacy of the BP/Au nanocomposites against E. faecalis (Fig. 13b). For more examples of nanomaterials beyond graphene (NBG), the reader is directed to an excellent review by Yin and Gu.
5. 3-D and other high-level nanomaterials

Compared to 0-D, 1-D, and 2-D nanomaterials, systems with a comprehensive 3-D or 3-D-like structure are only beginning to be used for biomedical applications. 3-D nanomaterials are materials that are not confined to the nanoscale in any dimension, these include nanoballs (dendritic structures), nanocoils, nanoclusters, nanocones, nanopillars and nanoflowers.\(^{139,140}\)

In addition, 3-D-like nanomaterials can be formed by the assembly of 0-D, 1-D, and 2-D nanomaterials. Moreover, there are some highly-complex composite materials with advanced supramolecular structure, which cannot be defined as 0-D, 1-D, or 2-D materials; so, they are also included and discussed here.

Two of the most prominent skeleton-like nanomaterials are metal–organic frameworks (MOFs) and covalent organic frameworks (COFs), which are generally biocompatible towards tissues.\(^{141,142}\) These are porous materials formed through the self-assembly of covalent bonds (COFs) or through the coordination between metal ions and multidentate ligands (MOFs). While both 2-D structured and 3-D structured MOFs and COFs are available, in particular 3-D structured ones (3-D MOFs/COFs) exhibit considerable antibacterial activity.\(^{141,142}\) In 2020, Qu and co-workers reported the construction of a MOF@COF hybrid that exhibited nanzyme characteristics (peroxidase-based) for the treatment of bacteria. This was the first example where COFs were used for tuning the catalytic and therapeutic performance of MOFs, resulting in the effective treatment of both *E. coli* and *S. aureus*.\(^{143}\) Mesoporous-based materials have been used to form fine microstructures. Examples include a mesostructured, Ag containing, silica-based calcium phosphate (80SiO2–CaO–P2O5 + Ag) and bioactive ceramic powders (Ag/80S) reported by Shih and co-workers.\(^{144}\) With the help of Ag as a therapeutic agent and the outside shell as a carrier, Ag/80S was shown to inhibit MDR bacteria, while acting as a bone graft material with osteoinductive and osteoconductive properties. In addition, Peng and co-workers developed an antibacterial polymer based on mesoporous bioactive glass (MBG) for bone material applications.\(^{145}\) MBG was first modified with dopamine, which underwent oxidative self-polymerization to form pMBG, which bound and reduced Ag\(^+\) ions to metallic Ag (Fig. 14a). Ag@pMBG was incorporated into a polymer scaffold (PLLA-PGA) to provide long-term antibacterial activity against *E. coli* with good cytocompatibility, which can only be ensured with the presence of incorporated Ag (Fig. 14b).

A novel “nanocube” was developed by Liu and co-workers, where a zeolitic imidazolate framework (ZIF-8) was pyrolyzed at 800 °C to form an NCS (nanocube-like structure) followed by the adhesion of silver ions to the surface of the NCS by sonication in ethanol.\(^{146}\) With the addition of sulphur, the metal ions can in situ form small metal chalcogenides on the surface of the NCSs. Both PDT and PTT effects were observed with good NIR response and low cytotoxicity. *S. aureus* was effectively treated using excitation by an 808 nm laser for 20 min. A TiO\(_2\) nanorod array was developed by Zhang and co-workers in 2021, with multiple mechanisms including PDT, PTT, and physical killing to produce excellent antibacterial properties on titanium towards *E. coli* and *S. aureus* after 15 min of 808 nm irradiation.\(^{147}\) The system was applied for *in vivo* antibacterial treatment including in mouse tissue infection models and the bone tissue around the Ti implants. Moreover, this nanorod array was found to improve new bone formation around implants.

Ag-decorated polydopamine/mesoporous silica composites were constructed to enhance the therapeutic efficiency of Ag and reduce toxicity via an incorporating/releasing effect triggered by pH changes and ROS, and was used for the treatment of drug-resistant bacteria and tumor cells.\(^{148}\) Mesoporous cellular silica foams (MCF) were demonstrated by Xia and co-workers as excellent antibacterial hemostatic agents.\(^{149}\) Metallic glass consisting of Mg, Ag, and Cu has been reported by Wang and co-workers for the long-term treatment of *S. aureus* and *E. coli* with the sustainable release of Cu and Ag ions.\(^{150}\) Compared with all the individual components, this system was shown to perform significantly better due to synergistic effects. Other strategies include the development of 3-D-based graphene derivatives for antibacterial applications. For example, tannic acid was used to reduce GO and induce self-assembly to form a graphene hydrogel,\(^{151}\) which represents an environmentally-friendly, and cost effective approach to treat both *S. aureus* and *E. coli*. The 3-D graphene exhibited high porosity, low density, hydrophobicity, good mechanical performance and thermal stability compared with normal 2-D graphene, with the ability to adsorb a range of oils, dyes and organic solvents. These results exhibited promise for water purification applications with the ability to treat *S. aureus* and *E. coli*.\(^{152}\) Bahadur and co-workers developed a 3-D layered double nanohybrid consisting of Mg–Al layered double hydroxide-reduced GO (rGO). The
synergistic antibacterial effect observed against E. coli was much better than those for the individual components with enhanced selectivity, which was attributed to protein degradation and GSH loss through the induction of oxidative stress.\textsuperscript{153} Moreover, a vertical heterostructure consisting of MoS\textsubscript{2}–coated rGO was developed by Yu and co-workers using a microwave-assisted hydrothermal method, which exhibited in situ bacterial binding with enhanced nanomotive activity and producing ROS for excellent E. coli/ S. aureus antibacterial effect.\textsuperscript{154} The overall effects observed within the heterostructure were synergistically improved compared with those of single MoS\textsubscript{2} or rGO. The mechanism was relevant for the catalytic generation of a range of ROS including hydroxyl radicals (OH\textsuperscript{•}) from H\textsubscript{2}O\textsubscript{2} by the nanozyme upon light activation, making the system suitable for use in mouse wound-infection models.

Huang and co-workers reported a degradable 3-D-printed polylactic acid (PLA) scaffold for applications in bone tissue engineering.\textsuperscript{155} The PLA scaffold was elaborated around a 3-D-printed core functionalized with an adherent polydopamine coating, and the modified scaffold was then used to immobilize a mixture of gelatin, needle-like nanohydroxyapatite (nHA), and ponericin (Fig. 15a). Long-term antibacterial activity was observed towards E. coli and S. aureus (Fig. 15b). Zheng and co-workers developed AgNP-associated carbon aerogels p-BC/AgNPs for antibacterial applications.\textsuperscript{156} p-BC/AgNPs exhibited excellent antibacterial efficiency against E. coli and S. aureus, which was better than with just AgNPs and p-BC. Moreover, p-BC/AgNPs exhibited excellent cell attachment and biocompatibility. Due to these properties, the authors proposed that this newly developed antibacterial material could be used for wound dressings, medical implants, and drug release applications.

Conclusions

Bacterial infections are a major health concern, threatening the lives of millions of people worldwide. Without effective treatments, a wide range of serious health complications will develop, including high mortality rates.\textsuperscript{3} The emergence of MDR “superbugs” is now becoming more and more problematic in clinical scenarios. The use of functional materials represents an exciting opportunity for bacterial treatments due to their outstanding PDT/PTT properties and drug carrier (loading/releasing) properties, which provides an opportunity to reduce the risks of developing drug-resistant bacteria.\textsuperscript{157} Currently, the major focus of nanomaterials research has been towards anticancer applications. The antibacterial applications of nanomaterials started a decade ago, using them as functional building blocks for the fabrication of antibacterial surfaces.\textsuperscript{158,159} Graphene-based antibacterial papers were initially developed where macroscopic GO and rGO papers can be conveniently fabricated from their suspension through simple vacuum filtration, and were found to inhibit the growth of E. coli.\textsuperscript{160} A new approach to fabricate antibacterial cotton fabrics by attaching single layer GO has also been reported, and the properties of GO remained even after repeated washing (ca. 100 times). Appropriate levels of crosslinking can be induced both by radiation and by chemical methods. Compared with single cotton or GO, the composite system formed by cotton and GO has an improved antibacterial effect towards E. coli and B. subtilis.\textsuperscript{161} In this review, we provide an insight into recent research that has focused on the use of functional nanomaterials for the treatment of bacteria. The research covered in this review has been organised according to the materials dimensions, clearly highlighting the importance of the morphologies of the nanomaterials used.

Despite the many successful experiments demonstrating the use of these systems, their use for clinical applications still represents a major hurdle due to their instability and potential side-toxicity in vivo. As such this has significantly prevented their development into commercially available therapeutics, which is further exemplified by the scarcity of functional materials being evaluated in vivo.\textsuperscript{162} However, the most recent in vivo studies have demonstrated excellent therapeutic efficacy without any serious toxicity being observed. Possible methods for enhancing biocompatibility may include functionalisation of the nanomaterial platforms with carbohydrates and peptides to afford targeted systems. Furthermore, these biomolecules may provide additional antibacterial properties. A representative example is polymeric antimicrobial peptides (AMPs) that have been recently developed by Liu and co-workers,\textsuperscript{163–165} which are cationic\textsuperscript{164} and amphiphilic\textsuperscript{165} and exhibit antifungal activity,\textsuperscript{163} immunomodulatory functions,\textsuperscript{165} and significant time-resolved effects against multiple bacterial species and biofilms both in vivo and for mouse infection models. In the near future, nanomaterials emerging from the developments in materials science, will result in improved clinically relevant properties such as stability, biocompatibility, and biodegradability, and result in more tissue-friendly targeting/assisting agents. With this review we have summarized the current state of the art for nanomaterial-based antibacterial systems and provided guidelines for the development of new and improved nanomaterial platforms. We are confident that with sustained hard work in this area,\textsuperscript{166}
an antibacterial nanomaterial system could soon be available to treat patients with MDR infections.

**Abbreviations**

**E. coli**  
*Escherichia coli*

**S. aureus**  
*Staphylococcus aureus*

**MRSA**  
Methicillin-resistant *S. aureus*

**P. aeruginosa**  
*Pseudomonas aeruginosa*

**S. epidermidis**  
*Staphylococcus epidermidis*

**P. gingivalis**  
*Porphyromonas gingivalis*

**P. micros**  
*Peptostreptococcus micros*

**P. intermedia**  
*Prevotella intermedia*

**S. mutans**  
*Streptococcus mutans*

**B. subtilis**  
*Bacillus subtilis*

**A. baumannii**  
*Acinetobacter baumannii*

**S. faecalis**  
*Streptococcus faecalis*

**E. cloacae**  
*Enterobacter cloacae*

**B. pertussis**  
*Bordetella pertussis*

**K. pneumoniae**  
*Klebsiella pneumoniae*

**P. mirabilis**  
*Proteus mirabilis*

**S. marcescens**  
*Serratia marcescens*

**DDS**  
Drug delivery system

**PDT**  
Photodynamic therapy

**PTT**  
Photothermal therapy

**Ag**  
Silver

**Ti**  
Titanium

**Cu**  
Copper

**Pd**  
 Palladium

**Se**  
Selenium

**ROS**  
Reactive oxygen species

**0-D**  
Zero-dimensional

**1-D**  
One-dimensional

**2-D**  
Two-dimensional

**3-D**  
Three-dimensional

**GQDs**  
Graphene quantum dots

**AgNps**  
Silver nanoparticles

**NCC**  
Nanocrystalline cellulose

**ν-Cys-AgNPs**  
ν-Cysteine functionalised AgNPs

**EPS**  
Extracellular polymeric substances

**ν-Cys**  
ν-Cysteine

**Bi**  
Bismuth

**BiNPs**  
Bismuth nanoparticles

**γ-CD**  
γ-Cyclodextrin

**MOFs**  
Metal–organic frameworks

**GRGDs**  
Gly-Arg-Gly-Asp-Ser

**PVA**  
Poly(vinyl alcohol)

**CNDs**  
Carbon nanodots

**OD**  
Optical density

**MC3T3-E1 cells**  
Mouse embryo osteoblast precursor cells

**CA**  
Citric acid

**CDs**  
Carbon dots

**GO**  
Graphene oxide

**TEM**  
Transmission electron microscopy

**SEM**  
Scanning electron microscopy

**AuNPs**  
Gold nanoparticles

**DAPT**  
4,6-diamino-2-pyrimidinethiol

**DGNPs**  
(4,6-Diamino-2-pyrimidinethiol)-modified gold nanoparticles

**uDGNPs**  
Ultrasmall DGNPs

**MOF@CeO₂NPs**  
CeO₂-decorated nanoparticle MOFs

**ATP**  
Adenosine triphosphate

**Ce**  
Cerium

**Zr**  
Zirconium

**MEH-PPV**  
2-Methoxy-5-((2-ethylhexyl)oxy)-p-phenylenevinylene

**BFPB**  
4-[2,2-Bis[4-formylphenoxy]methyl]-3-[4-formylphenoxy]propoxy]benzaldehyde

**TMB**  
3,3',5,5'-Tetramethylbenzidine

**CEMA**  
2-Chloroethyl methacrylate

**HEMA**  
Hydroxyethyl methacrylate

**SWNTs**  
Single-walled nanotubes

**MWCNTs**  
Multi-walled carbon nanotubes

**PES**  
Polyethersulfone

**UF**  
Ultrafiltration

**GNRs**  
Graphene nanoribbons

**PEO**  
Polyethylene oxide

**NIR**  
Near-infrared

**PP4N**  
Polycationic porphyrin

**4MPFG**  
N,N,N',N'-Bis(pyridyl-4-methyl)-N-fluorenyl-9-methoxycarbonyl (Fmoc)-L-glutamate

**HBTC**  
1,3,5-Benzene-tricarboxylic acid

**AgNP–NA–rGO**  
AgNP/sodium 1-naphthalenesulfonate-functionalized reduced graphene oxide

**AQS**  
Anthraquinone-2-sulfonate

**PEG**  
Polyethylene glycol

**MoS₂**  
Molybdenum disulfide

**HT**  
Holotomographic

**MDR**  
Multidrug-resistance

**QAS**  
Quaternary ammonium salt

**NBG**  
Nanomaterials beyond graphene

**rGO**  
Reduced GO

**AgNP–NA–rGO**  
AgNP/sodium 1-naphthalenesulfonate-functionalized reduced graphene oxide

**AQS**  
Anthraquinone-2-sulfonate

**NF**  
Nitric oxide

**RNS**  
 Reactive nitrogen species

**ONOO⁻**  
 Peroxynitrite

**BNN6**  
N,N'-Di-sec-butyl-N,N'-dinitroso-1,4-phenylenediamine

**Lys-MoS₂**  
Lysozyme-coated MoS₂ nanosheets

**VLP**  
Polyvinyl pyrrolidone

**WS₂**  
Tungsten disulfide

**BP**  
Black phosphorus

**COPs**  
Covalent organic frameworks

**MBG**  
Mesoporous bioactive glass
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